Final Report

SCIENTIFIC PEER-REVIEW OF THE HgCAMx ATMOSPHERIC MERCURY MODEL AND ITS APPLICATION TO THE 2002 ANNUAL CYCLE

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16 April 2004

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1.0 INTRODUCTION

Mercury pollution is a growing concern worldwide. In the U.S. mercury renders more surface waters impaired for fishing than any other toxic contaminant and fish consumption is often the most significant route of human exposure to mercury. Consequently, the U.S. Food and Drug Administration routinely issues fish consumption guidelines that are complimented in many states by mercury fish advisories. The Great Lakes Region is no exception. In response to this environmental threat to air, water, terrestrial resources and portions of the food supply, several science groups in the U.S. have been actively conducting mercury modeling research for several years. These include the Electric Power Research Institute (EPRI), the U.S. EPA, and the Wisconsin Department of Natural Resources (WDNR).

Recognizing the importance of atmospheric mercury, it's deposition to land and water bodies in the Great Lakes Region, and subsequent entry into the food chain, the WDNR has been a leader in research into the environmental impacts of mercury (Hg) pollution. Since the 1970s the Department has supported work to address this problem in a number of different ways. Recently, aided by an EPA grant and WDNR in-kind support funds, the Department sponsored the development of a new regional-scale atmospheric mercury modeling system for the Great Lakes region. This new model, known as HgCAMx, is destined to play an important role in evaluating strategies to reduce the amount of mercury emitted by coal-burning power plants and other major emitters in the Great Lakes region.

Consistent with WDNR's overall atmospheric mercury modeling system program plan, the Department commissioned an independent scientific peer-review of the HgCAMx modeling system to complement the model develop and evaluation process. Our methodology for reviewing the WDNR mercury modeling system focused on the most important chemical and physical processes; owing to inherent resource limitations, less emphasis was placed on processes that are known or suspected to have little influence on model predictions. Very little attention was given to the details of the model coding or logic. To optimize the resources available for this review, we attempted to build upon previous scientific peer-reviews of the various emissions, meteorological, and air quality system components of the model (e.g., Tesche et al., 1992; Kumar and Lurmann, 1997; Roth et al., 1998) and the science algorithms adopted from other models and included in the HgCAMx system (e.g., ENSR, 1993; AER, 1996).

1.1 Summary of Mercury Pollution

All forms of mercury can have some effect on the human nervous system, although the form that is most responsible for human health impacts is methylmercury. This is also the form of mercury found in fish. Methylmercury is a neurotoxin that causes brain and nervous system damage. Those most at risk from methylmercury are children and unborn babies of mothers who eat mercury-contaminated fish during pregnancy. Harmful concentrations of mercury in fish in a particular water body can come from multiple sources. In order to adequately reduce mercury concentrations in fish in a particular water body, the relative importance of the different sources of the mercury (i.e., other water bodies, land, direct discharges, and wet and dry atmospheric deposition) needs to be determined. Where atmospheric deposition of mercury is a major contributor to contamination in a particular water body, then the relative importance of local, regional, and even global sources of mercury become an important consideration.

Mercury exists in trace amounts in fossil fuels (e.g., natural gas, oil, and coal), vegetation, crustal material, and waste products. Through combustion or natural processes, mercury vapor is released to the atmosphere, where it can drift for a year or more, spreading across vast regions of the globe. Roughly 4900 tons of mercury are emitted annually into the atmosphere worldwide from both natural and anthropogenic sources, while in the U.S. coal-fired power plants release approximately 48 tons of mercury

per year (Feeley et al., 2003). While this amounts to about 1% of the global emissions rate, mercury emissions from power plants can be much more significant in regions immediately downwind of the EGUs (e.g., the Great Lakes).

Mercury is emitted from coal-fired utility stacks as either gaseous elemental mercury, Hg(0), gaseous inorganic oxidized mercury, Hg(II), or particulate mercury, Hg(p). In the atmosphere, Hg(0) is the dominant form (Schroeder and Munthe, 1998). Hg(II) and Hg(p) constitute typically only a few percent of total mercury. Hg(0) is only slightly soluble and is not removed significantly by wet deposition; its dry deposition velocity is also believed to be low. Consequently, Hg(0) has an atmospheric lifetime on the order of several months that is governed by its oxidation to Hg(II). In contrast, Hg(II) is quite soluble and is removed rapidly by wet and dry deposition processes. Subsequently, Hg(II) may become methylated in water bodies and bioaccumulate in the food chain. Particulate mercury, Hg(p), is mostly present in the fine fraction of particulate matter ($PM_{2.5}$), although some Hg(p) may be present in coarse PM (e.g., Landis and Keeler, 2002).

Modeling atmospheric mercury deposition is complex because it involves the emissions, transport, turbulent dispersion, chemistry and deposition of mercury over spatial scales ranging from local to global. Given the wide range of time and space scales involved and the many competing oxidation-reduction pathways involving natural and manmade sources, it is not surprising that many different models have been developed both in the U.S. and abroad, particularly in Europe (Pirrone, 2001). Indeed, Ryaboshapko and co-workers (2002) report on international research efforts to apply and evaluate five (5) contemporary atmospheric mercury chemistry models using a common episodic data base. This review paper and the references contained therein provide a good summary of mercury modeling activities through 2001. Since that time, additional mercury modeling work has been carried out by many organizations.

1.2 WDNR Mercury Model Development and Evaluation

The HgCAMx model development was a collaborative effort between WDNR technical staff, other state modeling groups within the Midwest Regional Planning Organization (MRPO), and private sector modeling contractors. The result has been a new state-of-science atmospheric mercury modeling system--HgCAMx. As described by Yarwood et al. (2003a), initial model development activities have been completed and the code was subsequently evaluated with region-specific data for the full year 2002 annual cycle.

The HgCAMx model was developed using the CAMx4 (ver 4.02) regional ozone/fine particulate model as the host system (See ENVIRON [2003] for details on the CAMx modeling system). The TEAM mercury chemistry module, previously tested and evaluated by Seigneur et al. (2001a, 2003b), was implemented in CAMx4 by scientists at Atmospheric & Environmental Research, Inc. (AER). The initial full-scale test of the HgCAMx model involved the development of an annual 2002 modeling database at 36 km horizontal resolution over the "National RPO grid" covering the entire continental United States and parts of Canada and Mexico. As noted, several groups were active in developing the 2002 HgCAMx modeling data base. WDNR performed annual MM5 meteorological modeling to develop the 2002 meteorological files and the agency also assembled the mercury emissions portion of the annual modeling inventory. For the ozone and fine particulate precursor species (e.g., VOC, NO_x, CO, SO_2, crustal material, soil) the Lake Michigan Air Directors Consortium (LADCo) developed emission databases, primarily in support of ongoing Midwest RPO studies. Mercury boundary conditions were constructed from global model simulations by AER while ENVIRON constructed the other HgCAMx model inputs and carried out the model evaluations. The report by Yarwood et al. (2003a) represents the most up to date summary of the HgCAMx model development and initial performance testing available.

1.3 Objectives of the HgCAMx Peer Review

The goal of this study was to conduct an independent technical review of the HgCAMx model, its supporting emissions and meteorological inputs, and the process whereby the overall modeling system (emissions, meteorology, chemical transport, transformation and removal) was evaluated with available meteorological and air quality data in the study region. Specific objectives of our peer-review included:

> Assess the adequacy of the emissions inventory of mercury sources including appropriate emission estimates, missing categories of sources, spatial distribution, temporal profiles, and the use of appropriate speciation profiles;

> Assess the application and evaluation of the MM5 meteorological model as a preprocessor of meteorological parameters for annual operation of the HgCAMx model;

> Assess the scientific formulation, technical implementation, and practical operation of the HgCAMx atmospheric model including appropriate mercury chemistry and physical processes and chemical and physical processes related to ozone and particles;

> Assess the adequacy and findings of the HgCAMx model performance evaluation (Yarwood et al., 2003a) carried out at the completion of the model development program; and

> Provide recommendations for improvements to the HgCAMx modeling system and/or its attendant application methodologies;

The time and resources available to carry out this study required that our review of the HgCAMx model, its input data bases and formal performance evaluation be focused on the most important components of this new modeling system. Independent peer-review is an important element of any new model development activity, and we have attempted to cover the most important components of the HgCAMx system first. Peer-review is an ongoing process as the emergent modeling system is used by an ever widening group of modelers and applied to a broader set of atmospheric and regulatory conditions. Even mature models such as CAMx (the host framework for HgCAMx) undergo continuing peer-review and refinement as greater experience is gained in using the model and in applying it to more episodes and physical domains.

It is important to recognize that many components of the newly-assembled HgCAMx modeling system (EMS-2003, MM5, and CAMx) have already undergone substantial peer-review by numerous groups in the course of typical applications. However, even so-called mature modeling systems such as EMS-2003, MM5, or CAMx are occasionally found to contain coding or logic errors or other inconsistencies as the model is adapted to new conditions or exercised by new users. Coding errors are common in all new models and only rigorous testing and practical application by many groups can reduce the number and importance of such errors. The HgCAMx code has already been subjected to a battery of tests by the developer (ENVIRON) to identify and correct errors. Once the initial model development and code verification has been completed by the developer, identification of residual coding errors and logic problems becomes a very difficult and time consuming task. It is certainly beyond the scope of this initial peer review. Accordingly, we have focused on a higher tier of issues such as adequacy and completeness of the modeling inputs data sets, model formulation and operation, the initial performance assessment, and known areas where model difficulties or performance improvement opportunities exist.

Some potential shortcomings or limitations of the current version of the HgCAMx model are already known to the WDNR and the model development team. Some of these, identified by the developers (Yarwood et al., 2003a) may not have known, easy or practical solutions. Thus, as part of this review, we have attempted to identify possible areas of improvement for known or suspected problems.

1.4 Report Structure

This report consists of seven Chapters. In the following Chapter 2, we discuss the overall HgCAMx model formulation against the backdrop of other contemporary mercury modeling systems worldwide and in consideration of the particularly important chemical and physical processes that a govern the fate of Tropospheric mercury over regional and continental scales. Brief comments are provided regarding the current model's computer architecture and coding. In Chapter 3 we review the basic emissions inventory data bases for the various precursors species (VOCs, NOx, Hg) important in regional-scale mercury modeling. The processing of these national inventories with the EMS-2003 emissions modeling system is addressed in Chapter 4 with the aim of evaluating the adequacy of the methods used to generate model-ready emissions data sets for simulation of the 2001 annual cycle. The development and evaluation the meteorological inputs to HgCAMx, produced via application of the MM5 prognostic meteorological model, is the subject of Chapter 5. In this chapter, we also report our own independent evaluation of the MM5 modeling system to shed further light on the model's performance relative to other recent regulatory modeling studies using MM5 of comparable models. Chapter 6 focuses on the existing performance of the HgCAMx system as reported by Yarwood et al., (2003a) with the aim of assessing independently the findings of this initial evaluation. We also suggest subsequent evaluation steps that the WDNR may wish to carry out in order to develop an even better understanding of the modeling systems current performance capabilities and areas for further model refinement. Our findings and recommendations are presented in Chapter 7.

2.0 HgCAMx MODEL FORMULATION REVIEW AND ASSESSMENT

This chapter discusses the formulation of the HgCAMx mercury model in the context of the key chemical and physical processes known or believed to govern the fate of mercury in its various forms over regional scales for periods of a few hours up to a year or more. The model is also examined relative to other numerical models developed for atmospheric mercury, both in the U.S. and abroad. Particular emphasis is placed on the models formulation of transport, transformation and removal processes. We also discuss the coding of HgCAMx and the extent of current technical and user documentation for the model.

Consistent with the scope of this peer review, we focus on the HgCAMx model. However, a technical assessment of the model cannot be made absent knowledge of other contemporary regional and global scale mercury modeling tools. Since this study was not intended to yield a comprehensive comparison of atmospheric mercury models world-wide, we have relied on the excellent four-stage intercomparison study currently nearing completion (Ryaboshapko et al., 2001; 2002; 2003.) This project, organized by the Meteorological Synthesizing Center-East (MSC-E), is being performed by scientists from several countries, including some who have had direct involvement in the development, testing, and/or evaluation of key algorithms in HgCAMx or its predecessor models. The reader is referred to the above mentioned research project reports for an in-depth description and intercomparison of various atmospheric mercury model attributes and performance results.

2.1 Atmospheric Processes Relevant to Mercury Modeling

Mercury is emitted into the atmosphere from both anthropogenic (e.g., fossil fuel combustion) and natural (e.g., volcanic or geothermal) processes and from re-entrainment of deposited materials. Mercury is emitted in three forms: elemental mercury vapor, reactive (i.e., oxidized) gaseous mercury, and particulate mercury. One of the largest uncertainties with respect to anthropogenic sources of mercury is the speciation of the emissions into elemental, oxidized, and particulate components. In addition, natural sources such as forest fires may also be important but poorly quantified sources of mercury.

Once introduced into the atmosphere, these three forms are transported, chemically transformed, and then deposited back to the earth's surface at various rates. The rates of transport and transformation depend upon the chemical form of the emitted mercury, the height of the emissions, and the atmospheric conditions (e.g., wind speeds, precipitation, composition of oxidizing and reducing species). Elemental mercury emitted high above the ground can persist in the atmosphere up to a year or more[1] and be transported around the world while the more reactive forms can be deposited to land or water surfaces much closer to the source(s). Airborne mercury can also undergo chemical reactions that lead to mercury compounds that are more quickly deposited to the earth's surface.

Mercury transformation processes include gas-phase reactions, reactions in the aqueous phase (e.g., cloud droplets), gas/aerosol physical and chemical processes, and aerosol (i.e. particle) processes. The gas-phase reactions include the oxidation of elemental mercury to oxidized mercury by ozone, hydrogen chloride, hydrogen peroxide and molecular chlorine. The aqueous-phase chemistry includes the reduction of oxidized mercury to elemental mercury via reaction with hydroperoxy radicals and by the formation of sulfite complexes as well as the oxidation of elemental mercury by dissolved ozone, hydroxyl radicals and chlorine. Gaseous oxidized mercury can also be adsorbed onto particulate matter.

[1] Recent research by Hedgecock and Pirrone (2004), however, suggest that the lifetime (τ) of elemental mercury may be as short as 10 days under certain conditions; these researchers suggest that the atmospheric residence time of gasoues mercury may be much less than the generally accepted residence time of a year or more.

The fate of mercury in the atmosphere strongly depends on the transport and transformation processes. Key uncertainties include the completeness of the assumed aqueous phase mercury conversion processes and the amount of mercury gas that can adsorb onto particles. The temperature dependence of many reactions are not yet known and the conversion processes depend upon concentrations of other air pollutants that are known with varying degrees of certainty.

Airborne mercury reaches the land and water via wet and dry deposition processes. All three major forms of mercury can be dry deposited and the rates of deposition will vary depending upon surface characteristics and meteorological conditions. Wet deposition rates depend on cloud composition and precipitation patterns. Scavenging of mercury by precipitation varies considerably. Little if any scavenging occurs for elemental mercury while all of the oxidized form is likely to be scavenged. Roughly half of the particulate form of mercury may be scavenged (e.g., Seigneur et al., 2003a).

It is important to grasp the key roles that other atmospheric species play in the transformation and deposition processes. As noted, ozone, sulfur compounds, and particulates are particularly important in mercury atmospheric chemistry. This means that emissions from several other important air pollutants directly affect the fate of mercury in the atmosphere. These include emissions of nitrogen oxides and volatile organic compounds which form ozone as well as secondary aerosol particulates, emissions of sulfur dioxides which contribute to particulate and other gaseous sulfur compounds, and direct emissions of particulate matter. All of these other chemicals, well-known for their own harmful impacts on human health and the environment, have been incorporated into emergent, state-of-science 'one-atmosphere' models such as HgCAMx and EPA's Models-3/CMAQ.

2.2 Overview of Atmospheric Mercury Modeling Methods

In the U.S., the leading science groups spearheading mercury modeling research include the Electric Power Research Institute (EPRI), the U.S. EPA, and the Wisconsin DNR. EPRI has been active in supporting the development of the TEAM model (Seigneur et al., 2000, 2003a,b) while EPA has implemented mercury chemistry into the Models-3/CMAQ system (Bullock and Breheme, 2002). Mercury chemistry modules have been incorporated into other regional-scale Eulerian models including ADOM (Peterson et al., 2001) and SAQM (Xu et al., 2000a,b). Abroad, vigorous mercury model development abroad has also occurred and Ryaboshapko et al., (2002) chronicle recent model development and evaluation activities with several European and U.S. models. The various modeling frameworks differ in many respects, reflecting no doubt the predilections of the model developers, the research or applications challenge at hand, the availability of data bases and supporting modeling and computational facilities, and other factors.

The 'box' concept has all been used to construct several atmospheric mercury models, principally in western Europe. For example, the Chemistry of Atmospheric Mercury (CAM) model is a box model that contained (at the time of its development) a comprehensive chemical mechanism including 180 gas phase and aqueous phase chemical reactions involving more than 90 species (Pleijel and Munthe, 1995). CAM treats the details of VOC and NO_x photochemistry leading to the formation of ozone and HO_x. The model treats four gas-phase mercury species and eight aqueous-phase Hg species absorbing to soot particles within droplets. CAM also treats particulate within droplets as well as Cl_2 and SO_x chemistry. The MSC-E HM is another mercury box model but this code is derived from the grid-based TCM model (Petersen et al., 2001) described below. MSC-E HM includes three gas-phase Hg species, one particulate species, and six aqueous phase species and also three mercury species absorbed to soot particles within droplets.

In the grid-based model class, the Tropospheric Chemistry Model (TCM) is a three-dimensional chemical transport model based on the ADOM code (Petersen et al., 2001) developed in the U.S. and Canada a decade ago. The chemistry of TCM is a simplified version of the original chemistry package in the CAM box model described above. Here, detailed mercury chemistry has been sacrificed in order to allow the model to provide greater and more realistic representation of atmospheric transport processes. In the TCM model, convective cumulus and stratus cloud processes are modeled explicitly and both precipitating and non-precipitating clouds can be treated depending on the atmospheric conditions.

Another atmospheric chemistry model built upon a legacy regional photochemical model was reported by Xu et al., (2000a,b). These researchers implemented mercury chemistry into the SARMAP air quality model, a revised version of the RADM code applied to regional ozone air pollution in the California Central Valley in the early 1990s. A mercury mechanism containing the three key species (elemental, divalent, and particulate) were added to SAQM's CB4 mechanism and the model was refined to treat mercury processing in precipitating clouds, co-existing non-precipitating clouds and fair weather clouds. The model was tested with data in Connecticut for a summer week and a winter week.

The U.S. EPA has extended the Community Multiscale Air Quality Model (CMAQ), described by Byun and Ching, 1999) to include mercury chemistry. As described by Bullock and Brehme (2002), CMAQ Hg simulates transformations of mercury wit four new chemical reactions within the standard CMAQ gas-phase chemistry mechanism and a highly modified cloud chemistry module which includes compound-specific speciation for oxidized forms of Hg, seven new aqueous-phase Hg reactions, six aqueous Hg chemical equilibria, and a two-way mechanism for the sorption of dissolved oxidized Hg to elemental carbon particles. The model also simulates the partitioning of reactive gaseous mercury (RGM) between air and cloud water based on Henry's law for mercuric chloride. In CMAQ Hg, wet deposition is calculated based on the precipitation rate information produced by MM5 (the meteorological processor for CMAQ) in concert with the speciation of Hg in the cloud chemistry module. Dry deposition is based on CMAQ's standard deposition algorithm, involving the species-specific deposition velocity and the concentrations of the three forms of Hg near the ground.

A third comprehensive grid-based atmospheric mercury model was developed by Atmospheric and Environmental Research (AER) and the Electric Power Research Institute (EPRI). The Trace Element Analysis Model (TEAM) described by Pai et al., (1997), Seigneur et al., (2001a), Vijayaraghaven et al., (2003) is actually a part of a multi-scale modeling system that has been exercised from the global scale down to 20 km horizontal grid mesh (e.g., Vijayaraghaven et al., 2002). For regional to continental scale application, the TEAM model has been applied with six vertical layers extending from the ground up to an altitude of 6 km with layer interfaces at 60, 150, 450, 850, and 2000m. The TEAM mercury chemistry mechanism, originally described by Seigneur et al., (2001a) and later updated, includes the gas-phase oxidation of Hg(0) to Hg(II), the aqueous-phase oxidation of Hg(0) to Hg(II), the aqueous-phase reduction of Hg(II) to Hg(0), various aqueous-phase equilibria of Hg(II) species and the aqueous-phase adsorption of Hg(II) to PM.

In the TEAM modeling system, dry deposition is simulated in the conventional fashion using species-specific deposition velocities. A dry deposition velocity of 0.01 cm/s is used for Hg(0) while a value of 0.5 cm/s is used for Hg(II). As in CMAQ HG, the deposition rate in for each mercury species in TEAM is calculated as the product of the ground-level concentration in air and its dry deposition velocity. Wet deposition is estimated based on the species concentration, a precipitation scavenging coefficient, the frequency and speed of winds, and the fraction of time with rain. The scavenging coefficient of Hg(II) is estimated from its similarity with HNO_3. Since elemental mercury, Hg(0), has a very low solubility it is

assumed in TEAM to have a negligible wet deposition. In the TEAM applications reported in the literature (e.g., Pai et al., 1997; EPRI, 2000), the concentrations of O_3, SO_2, HCl, CL_2 and H_2O_2, and cloud water pH were estimated from measurements (i.e., not modeled with a 'one-atmosphere model) and the other gas-phase and aqueous-phase chemical species set to zero. This approach significantly reduces the computational burden of model such as CMAQ Hg and HgCAMx which include the full set of gas-phase photochemical and secondary aerosol reactions in order to calculate the concentrations of the various pollutant species that react with atmospheric mercury constituents.

Dastoor and Larocque (2004) describe a new real-time mercury transport/transformation model that has been coupled with Canada's operational Global Environmental Multiscale (GEM) model at the Canadian Meteorological Center. Designed as a comprehensive atmospheric global and regional scale mercury model, the GRAHM code is based on the gas phase and aqueous phase mercury chemistry model of Peterson et al., (1998, 2001), which is, as noted earlier, a simplified form of the Pleijel and Munthe, 1995 detailed mercury kinetic mechanism). Among the advantages of the GRAHM model is that is runs in lock-step with the GEM forecast model, providing near-real time forecasts of mercury air quality and deposition estimates. The model's formulation also reflects recent advances in boundary layer physics , cloud processing, and surface removal. However, reflecting its linkage to the Peterson and Pleijel and Munthe mechanism, the GRAHM modeling system does not treat the time and space variation in the various oxidant, SO_2, and particulate species that play key roles in mercury chemistry. These species are simply prescribed based on measurements and no information is available with which to assess the impact of these approximations on the reliability of the resultant mercury species predictions.

From this limited review, it is clear that modeling of atmospheric mercury has evolved along several lines, many reflecting the personal preferences of the model developers. Some models emphasize the treatment of chemical processes at the expense of being able to characterize physical atmospheric processes such as transport, turbulent diffusion, cloud-effects, and deposition processes. Other models address large (i.e., global) scales with a concomitant simplification in some physical or chemical processes. Other models treat the regional scale chemical transport and removal processes in depth, but have more limited ability to characterize the full body of knowledge regarding mercury chemistry and must make assumptions about the inflow boundary conditions. This latter limitation can be important in simulating mercury given the long atmospheric lifetimes of some Hg forms.

2.3 Formulation of the HgCAMx Model

2.3.1 Conceptual Formulation

HgCAMx is an extension of the current CAMx (mechanism 4) version of CAMx (ENVIRON, 2003) produced and distributed freely by ENVIRON International Corporation. The CAMx4 model version was augmented by the inclusion of the TEAM mercury chemistry package (Yarwood et al., 2003a). The CAMx4 version of the model was selected as the host framework largely because mercury has an affinity for particles that in turn influences its overall atmospheric chemistry. Recognizing the need to model atmospheric particles, the TEAM mercury chemistry was integrated with the full ozone and PM chemistry as represented in the CAMx4 model, yielding truly a 'one atmosphere' model. Chlorine also is an important species in mercury chemistry but present methods are insufficient to model it rigorously at this time. Except perhaps in Houston, gridded, diurnally varying chlorine emissions inventories are non-existent. Accordingly, in HgCAMx, prescribed chlorine (Cl_2, HCl) concentration levels are used based on day vs. night and land vs. water characterizations. While there is a research version of CAMx employing the CB4 kinetic mechanism extended for chlorine chemistry, this module was developed for simulating Cl_2 'hot spots' from industrial cooling towers (in Houston, TX). This specialized code is not yet suitable for simulating regional Cl_2 and/or HCl concentrations. To date, the HgCAMx model has been set up and

evaluated for a 2002 annual simulation on the national RPO grid at a 36 km horizontal grid scale. Thus far, the model evaluation has focused principally on the data from the national Mercury Deposition Network (MDN) data base.

As implemented for WDNR in the Great Lakes Region, the HgCAMx mercury modeling system consists of an emissions model (EMS-2003), a prognostic meteorological model (MM5), and an air quality or chemical transport model (HgCAMx). HgCAMx is formulated in general terms such that it can be exercised with alternative emissions (EPS2x, SMOKE) and meteorological (RAMS) preprocessor models. Below, we summarize key features of the HgCAMx model and provide our independent assessment of their reasonableness and adequacy.

2.3.2 Treatment of Transport Processes

While the HgCAMx model is capable of being operated with meteorology produced by either RAMS or MM5, the later model was chosen by WDNR as the processor for HgCAMx. This is a reasonable choice, since RAMS and MM5 have been shown to provide generally comparable performance characteristics in the Great Lakes Region (Tesche et al., 2001). MM5 is additionally attractive when one considers the unrestricted, public domain status of the code and the broader experience base with this model in regulatory studies. Since MM5 operates on the same Lambert Conformal map projection as CAMx4 and the EMS-2003 models, it is easy to integrate with these two meteorology-depend systems. Moreover, the choice of MM5 and the model application methodology used by WDNR staff in applying the model for the 2002 annual period is consistent with the approach being used by several states and Regional Planning Organizations (RPOs) for addressing regional haze and long term oxidant/fine particulate modeling. For addressing such issues, both the MM5 model and its application methodology have been successful in a number of ozone and fine particulate applications. However, accurate mercury deposition modeling places generally greater demands on the meteorological modeling system compared with an ozone or regional haze application owing to the much greater importance of cloud and wet and dry deposition processes.

WDNR set up the MM5 over the national RPO grid at 36 km scale (34 vertical layers) and carried out an annual simulation for use in HgCAMx model development and testing. Data from the National Center for Environmental Prediction (NCEP) was used for initialization and data assimilation. Science options included simple ice for moisture parameterization, the Kain-Fritch cumulus parameterization, the Pleim-Xiu PBL scheme and soil model, and the Rapid Radiative Transfer Model (RRTM) radiation scheme. Four dimensional data assimilation was used but only above the planetary boundary layer. We did not find an MM5 modeling protocol guiding WDNR's application of the model or any formal report summarizing the model application and performance evaluation results. However, the companion study by IDNR (Johnson, 2003) was of some use in assessing the present annual 2002 modeling for this application.

The HgCAMx final report includes a brief discussion of the MM5 set up and execution by WDNR staff, but there is little quantitative discussion of the model's performance. The developers note that the MM5 fields were tested with ENVIRON's METSTAT software and the WDNR "concluded that the MM5 produced reasonable meteorology fields that were suitable for annual air quality modeling". Yarwood et al., (2003a) also report that MM5 modelers at the Iowa DNR essentially replicated the WDNR's modeling for 2002 and obtained similar evaluation results.

As part of this peer-review, we performed an independent operational evaluation of the MM5 fields for the 2002 annual simulation and present the results in Chapter 5. As discussed there, we find that the

present characterization of transport in HgCAMx for the annual 2002 period is adequate for the purposed of the present model development and testing program.

The HgCAMx model developers (Yarwood et al., 2003a) indicate that a critical component for mercury deposition modeling is accurate prediction of precipitation. Indeed, as we saw in the Southern Appalachian Mountains Initiative (SAMI) project (Doty et al. 2002), unless the dynamic and thermodynamic fields are reasonably well simulated, it is unlikely that a grid-based air quality model will reproduce the correct amounts of cloud and precipitation and, correspondingly, wet deposition. Since a detailed examination of the adequacy of the MM5's precipitation modeling scheme was beyond the scope of this review, we focused instead on examining how the HgCAMx model utilized the MM5 modeled cloud and precipitation fields in developing estimates of wet deposition.

Adequate precipitation predictions are needed for credible mercury modeling. Indeed, this is one of the areas of model improvement identified by Yarwood et al., (2003a). They report sensitivity experiments focusing on the wet mercury deposition process and conclude that improved precipitation estimates from MM5 together with improved procedures in HgCAMx to use the precipitation fields to generate wet deposition warrant additional work. We concur with this assessment.

2.3.3 Treatment of Chemical Transformation Processes

The HgCAMx model consists of the integration of the "TEAM" mercury kinetic package into the CAMx-Mechanism 4 model, commonly referred to as CAMx4 and described in the most recent user's guide from the model developer (ENVIRON, 2003). The TEAM mercury mechanism is based on the work of Seigneur and co-workers (see for example, Seigneur et al., 2000, 2003a,b). Table 3-1 lists the current mercury mechanism as implemented in HgCAMx. Note that this mechanism is integrated with the regulatory version of the Carbon Bond IV (CB4) photochemical mechanism with isoprene updates. The mercury mechanism treats three Hg species, Hg(0), Hg(II), and Hg(p). Actually, Hg(II) consists of several chemical species in the gas phase and cloud droplets; Hg(II) can also adsorb to particulate matter. Since Hg(0) is relatively insoluable, wet deposition is only treated for Hg(II) and Hg(p) and is parameterized as the product of the cloud droplet concentration of the Hg species and the modeled precipitation amount. Dry deposition is treated using the resistance transfer approach of Wesley.

A key distinguishing feature of the HgCAMx model is the integration of the full ozone/PM chemistry with the gas-phase, aqueous-phase, and gas-liquid equilibria mercury chemistry in one consistent chemical mechanism. Because mercury concentrations are typically much smaller than the concentrations of active species in photochemical/aerosol reactions, the implementation of the TEAM chemistry into CAMx4 was straightforward. During the chemical integration time-steps, mercury species are assumed constant; at the end of the chemistry step, the mercury reactions are updated via algebraic expressions relating the various mercury species and the numerous oxidizing and reducing compounds (O_3, H_2O_2, OH, SO_2, and particulate matter) that influence the Hg chemical transformations. Owing to the lack of adequate emissions data on Cl_2 and HCl, these concentrations are prescribed rather than simulated within the chemical mechanism. Thus, unlike many other atmospheric mercury models (see Ryaboshapko et al., 2002), HgCAMx is truly a 'one atmosphere' model.

Beyond the inclusion of the mercury mechanism into CAMx4, the other main modifications to the host model included improvements to the dry deposition algorithms to address interannual variability and the effects of snow cover on deposition and photolysis rates. Yarwood et al., (2003a) provide a brief summary of these implementations.

Our brief review of the mercury mechanism incorporated into the HgCAMx model suggests that the present mechanism is a credible representation of the gas-phase and aqueous-phase reactions influencing atmospheric mercury given today's state-of-knowledge. Of course, this mechanism does not contain all known or speculate reaction pathways, due in part to the need to condense the reaction scheme to a manageable level. Unlike other models that neglect atmospheric transport pathways altogether or neglect the companion photochemical and secondary aerosol reaction mechanisms, the HgCAMx chemistry module is design to achieve an appropriate balance between computational speed and technical rigor given present knowledge. In our view, this balance has been achieved in a credible fashion. To be sure, ongoing research into the various mercury chemistry reactions, estimation or measurement of reaction rates, and improvements in the knowledge of gas-phase, aerosol, and aqueous-phase species interactions is continuing at a rapid pace. Accordingly, it is proper that the present mechanism not be viewed as static but should be updated as technically sound advancements become available. Fortunately, the flexible and modern architecture of the host CAMx model allows these mechanism refinements (and associated changes to the air quality model such as initial and boundary conditions, species lists, and so on) to be made in a straightforward manner.

2.3.4 Treatment of Removal Processes

Airborne mercury reaches the land and water via wet and dry deposition processes. All three major forms of mercury can be dry deposited and the rates of deposition will vary depending on the deposition surface characteristics and meteorological conditions. Wet deposition rates depend on the cloud composition and precipitation patterns. Scavenging of mercury by falling precipitation varies considerably. Not much if any scavenging occurs for the elemental mercury while all of the oxidized form is likely to be scavenged. Probably half of the particulate form of mercury is thought to be scavenged (Seigneur et al., 2002). Treatment of wet and dry deposition in the HgCAMx model is performed with algorithms that provide for coupling between the MM5 fields of temperature, wind, mixing and precipitation with the wet and dry deposition process descriptions embedded in the air quality model. We discuss dry and wet deposition separately.

Dry Deposition

The dry deposition algorithm for gases and aerosols in HgCAMx is based on the seminal development work by Wesley (1989) and has a number of conceptually similarities to the schemes used in several other regional models including the UAM-V, UAM-AERO, RADM and SAQM. In the Wesley scheme, the dry deposition velocity v_d is calculated from three primary resistances r (s/m) in series:

$$V_d = \frac{1}{r_a + r_b + r_s}$$

where the aerodynamic resistance r_a represents bulk transport through the lowest model layer by turbulent diffusion, the quasi-laminar sublayer (or boundary) resistance r_b represents molecular diffusion through the thin layer of air directly in contact with the particular surface to which material is being deposited, and the surface resistance r_s depends upon the properties of the surface and the depositing species. The HgCAMx development included two refinements to the Wesley scheme, namely:

> An improved the description of seasonal dependencies based on geographic location and time of year; and

> Specification of when surfaces are snow-covered and modified the dry deposition rates

accordingly.

The new HgCAMx seasonal map for dry deposition is a useful refinement in that it extends the Wesley methodology to provide a more continuous variation in the surface resistance estimates across the four seasons (by treating them monthly instead of seasonally) and by attempting to account for the variation in season definition by latitudinal bands.

In addition, the extension of the CAMx dry deposition formalism to include temporally and spatially varying snow cover is also a useful model enhancement. This refinement was included by defining an optional snow-cover data field in the albedo-haze-ozone column (AHO) file. Snow-covered grid cells can be prescribed in three different ways, the depending upon the nature of the modeling simulation, the region being modeled, and the availability of data. For the present model verification effort, the use of the NCEP ETA Data Analysis System (EDAS) data to define the time and space variation of surface snow cover was used. Grid cells were defined as snow-covered if the EDAS snow-cover fraction for the grid cell was greater than 0.5.

We believe that the updates made to the CAMx4 dry deposition routines for HgCAMx - the inclusion of temporally and spatially varying snow cover and seasonal variation in dry deposition surface resistances – are appropriate and reasonably implemented in the model.

Wet Deposition

Wet deposition is an important removal process for particles and relatively soluble gaseous pollutants and may occur via several alternative mechanisms including: (a) co-mixing of trace gases and condensed water, (b) adsorption of gas molecules by water droplets, (c) aqueous phase reactions of pollutants within water droplets, (d) precipitation of droplets to the ground or water surfaces, and (f) diffusion of ambient gases into falling solid or liquid precipitation. Since these steps may be reversible, the overall wet deposition rate depends on the net processes involved. In the CAMx host code, wet deposition is modeled with several different algorithms, depending upon the specific mechanism involved, i.e., the scavenging of ambient gases, scavenging of gases dissolved in cloud water, scavenging of in-cloud aerosols, or scavenging of dry particles. The basis scavenging concept in HgCAMx is that the local rate of change of concentration within or below a precipitating cloud depends upon a scavenging coefficient, Λ , And the concentration of the particulate or gaseous species. For dissolved species, in HgCAMx, the wet deposition flux is the product of its concentration in rainwater and the precipitation amount. Clearly if one or both components are misrepresented, or if the scavenging coefficient is poorly chosen, the net wet deposition rate will be incorrectly calculated.

Our review of the dry and wet deposition removal schemes in HgCAMx suggests that the present formulation is indeed a credible representation of the present state-of-knowledge of surface removal processes, as implemented in operational regional photochemical and secondary aerosol models. The treatment is far superior, both in terms of the number of processes represented and the detail given to each process, than the removal processes in older regulatory models such as the UAM (see, for example, the review by Tesche et al., 1992). Some other contemporary models employ somewhat different methods. CMAQ, for example, uses the RADM dry deposition scheme. When MM5 is run with the Pleim-Xiu LSM, there is an alternative Pleim dry deposition scheme that can be used with CMAQ that is not presently coded in CAMx. Presently, in the VISTAS Phase I modeling sensitivity study, these alternative deposition schemes (Wesley and RADM/Pleim) are being evaluated within a common host model (CMAQ) and results of this study may shed additional light on the subject. For the present, however, the Wesley scheme, as implement in HgCAMx appears to be reasonable.

We note, however, that while the RADM-based wet deposition treatment in HgCAMx seems reasonable, if the precipitation fields supplied to the model are seriously in error, incorrect wet deposition estimates may results. This indeed appears to have been the case in the annual 2002 model simulation. This issue is discussed in further in Chapters 5 and 6.

2.4 Model Code and Documentation

Resource constraints limited us from conducting a detailed examination of the HgCAMx source code. Typically, examination of a new model's source code line-by-line searching for logic or coding errors is tremendously resource intensive and generally not a wise use of time. Often coding difficulties are identified through actual model applications and diagnostic studies. However model developers have a battery of diagnostic and debugging tools available to them for testing new or modified code and these tools were employed by the ENVIRON model developers as part of the initial model building and verification effort.

The host CAMx model was developed using all new code during the late 1990s using modern and modular coding practices. Written entirely in ANSI standard FORTRAN 77, the model is highly modular in nature, allowing straightforward inclusion or modification of alternate science or computational routines. The code is written to permit compiler options (e.g., do/enddo loop definitions) that are commonly used on modern workstations and personal computers. Significantly, the model architecture does not include portions of legacy code held over from prior versions of the host model such as one finds in other grid-based air quality models in use today (e.g., UAM-V, REMSAD, URM, and MAQSIP). Unlike older models which represent a "patchwork" affair with considerable evidence of splicing of new features into older existing code or subroutines, CAMx4 presents a consistent level and style of documentation, punctuated by meaningful comments in the code, to allow users to follow the logic through the main program and subroutines.

We did independently set up the HgCAMx model code on Alpine's Linux Cluster running the Portland FORTRAN Compiler and encountered no difficulty in compiling the model and its numerous subroutines. However, resource constraints precluded us from actually exercising the HgCAMx modeling system using the 2002 input data sets developed by ENVIRON and the WDNR. But, in reviewing these data sets in support of other components of this peer review (see Chapters 3 through 5), we found no inconsistencies or difficulties from a computational standpoint.

The host CAMx4 modeling system is very well documented from both a science formulation and a model user's perspective (ENVIRON, 2003a). The CAMx4 modeling system may be downloaded without restriction from the model website (http://www.camx.com). This unrestricted public access of the HgCAMx model thus conforms explicitly EPA's requirements for models in regulatory decision-making (EPA, 1999; 2001). In addition to requiring that models used for regulatory decision-making be publicly available, EPA modeling guidance further states that users must be free to revise the code to perform diagnostic analysis" (EPA, 2001, pgs. 169-170.) ENVIRON's release of HgCAMx complies with the full spirit of this guidance. We note, however, that not all mercury models used in the U.S. comply with this requirement (e.g., REMSAD). WDNR's commitment to the HgCAMx modeling system being a truly public-domain model is commendable.

In addition to the formal documentation for HgCAMx, supplemental documentation on CAMx4 and HgCAMx may be obtained from recent technical papers and presentation given by the model developers (see, for example, Yarwood et al., 2003b; Morris et al., 2003a,b)

Some aspects of the HgCAMx code were not reviewed in detail owing to project focus and resource constraints. For example, a detailed examination of the new code would entail a review of the computer code and model documentation to ensure that each science and computational algorithm is coded consistent with the model's scientific formulation and underlying assumptions. This would be facilitated by installation of the modeling system on different computer platforms and replication of the 2002 annual test case to verify "transferability" (including getting the same answers) of the model code and data sets. Also, a focused line-by-line review of the new portions of the HgCAMx source code would lead to a better understanding of its structure and organization and would allow the reviewer to verify correct implementation of the science and computational algorithms. Other code-related investigations would include an analysis to examine the time sequencing of key operations in the chemistry and deposition routines to ensure accuracy and consistency and the performance of 'spot checks' of new code to search for: (a) undefined or un-initialized variables, (b) inconsistent specification or dimension of arrays common to multiple subroutines, (c) inconsistencies between subroutine calls and subroutine statements, and (d) other logical errors. Finally, the actual coding and operation of new preprocessor modules (e.g., snowcover) were not investigated but would certainly be appropriate. These types of model code verifications are time-consuming and resource intensive. The ENSR review of the UAM-V model (ENSR, 1993) in support of the Lake Michigan Ozone Study (LMOS) was the last time to our knowledge that such a detailed review was carried out for a grid-based photochemical model. The ENSR methodology provides a good example of how a detailed peer-review of an air quality modeling system might be carried out, given ample resources and time.

2.5 Synthesis of HgCAMx Model Formulation

Our review has not identified any significant potential or actual weaknesses in the HgCAMx model formulation. The mercury mechanism included in the CAMx4 host model is arguably the most comprehensive chemical transformation scheme yet incorporated into a regional chemical transport model for seasonal or annual use. Particularly noteworthy is the fact that HgCAMx does not rely on estimated or interpolated fields of atmospheric oxidants (e.g., OH, O_3, H_2O_2) or particulate matter for use in the chemical mechanism but simulates these photochemical and secondary aerosol precursor and produce species from first principals. Although this adds considerable computation burden to annual simulations, and the concomitant need to compile adequate precursor inventories for ozone and PM precursor species, it yields a truly one-atmosphere modeling system that accounts for the time and space variation of the key chemical species that participate in the fate of mercury in the regional atmosphere.

Table 2-1. Mercury Chemistry Treated in the HgCAMx Model.

Equilibrium Process or Chemical Reaction	Equilibrium or Rate Parameter[a]	Reference
$Hg(0)\ (g) \rightleftharpoons Hg(0)\ (aq)$	0.11 M atm^{-1}	Sanemasa, 1975; Clever et al., 1985
$HgCl_2\ (g) \rightleftharpoons HgCl_2\ (aq)$	1.4 x 10^6 M atm^{-1}	Lindqvist and Rohde, 1985
$Hg(OH)_2\ (g) \rightleftharpoons Hg(OH)_2\ (aq)$	1.2 x 10^4 M atm^{-1}	Lindqvist and Rohde, 1985
$O_3\ (g) \rightleftharpoons O_3\ (aq)$	1.13 x 10^{-2} M atm^{-1}	Kosak-Channing and Helz, 1983
$HCl\ (g) \rightleftharpoons HCl(aq)$	1.1 M atm^{-1}	Marsh and McElroy, 1985
$SO_2\ (g) \rightleftharpoons SO_2\ (aq)$	1.23 M atm^{-1}	Smith and Martrell, 1976
$Cl_2\ (g) \rightleftharpoons Cl_2\ (aq)$	0.076 M atm^{-1}	Lin and Pehkonen, 1998
$H_2O_2\ (g) \rightleftharpoons H_2O_2\ (aq)$	7.4 x 10^4 M atm^{-1}	Lind and Kok, 1986
$HgCl_2\ (aq) \rightleftharpoons Hg^{2+} + 2\ Cl^-$	10^{-14} M^2	Sillen and Martell, 1964
$Hg(OH)_2\ (aq) \rightleftharpoons Hg^{2+} + 2\ OH^-$	10^{-22} M^2	Sillen and Martell, 1964
$HCl\ (aq) \rightleftharpoons H^+ + Cl^-$	1.7 x 10^6 M	Marsh and McElroy, 1985
$Cl_2\ (aq) \rightleftharpoons HOCL + Cl^- + H^+$	5.0 x 10^{-4} M^2	Lin and Pehkonen, 1998
$HOCL \rightleftharpoons OCL^- + H^+$	3.2 x 10^{-8} M^2	Lin and Pehkonen, 1998
$SO_2(aq) + H_2O_2(aq) \rightleftharpoons SO_4^{2-} + 2H^+$	instantaneous [b]	McArdle and Hoffmann, 1983
$SO_2(aq) + H_2O(l) \rightleftharpoons HSO_3^- + H^+$	1.23 x 10^{-2} M	Smith and Martrell
$HSO_3^- \rightleftharpoons SO_3^{2-} + H^+$	6.6 x 10^{-8} M	Smith and Martrell
$Hg^{2+} + SO_3^{2-} \rightleftharpoons HgSO_3$	2.1 x 10^{13} M^{-1}	van Loon et al., 2001
$HgSO_3 + SO_3^{2-} \rightleftharpoons Hg(SO_3)_2^{2-}$	1.0 x 10^{10} M^{-1}	van Loon et al., 2001
$Hg(II)\ (aq) \rightleftharpoons Hg(II)\ (p)$	34 l/g	Seigneur et al., 1998
$Hg(0)\ (g) + O_3\ (g) \longrightarrow Hg(II)\ (g)$	3 x 10^{-20} cm^3 $molec^{-1}$ s^{-1}	Hall, 1995
$Hg(0)\ (g) + HCl(g) \longrightarrow HgCl_2(g)$	10^{-19} cm^3 $molec^{-1}$ s^{-1}	Hall and Bloom, 1993
$Hg(0)\ (g) + H_2O_2\ (g) \longrightarrow Hg(OH)_2\ (g)$	8.5 x 10^{-19} cm^3 $molec^{-1}s^{-1}$	Tokos et al., 1998
$Hg(0)\ (g) + Cl_2(g) \longrightarrow HgCl_2(g)$	2.6 x 10^{-18} cm^3 $molec^{-1}s^{-1}$	Ariya et al., 2002
$Hg(0)\ (aq) + O_3\ (aq) \longrightarrow Hg^{2+}$	4.7 x 10^7 M^{-1} s^{-1}	Munthe, 1992
$Hg(0)\ (aq) + OH\ (aq) \longrightarrow Hg^{2+}$	2.0 x 10^9 M^{-1} s^{-1}	Lin and Pehkonen, 1997
$HgSO_3\ (aq) \longrightarrow Hg(0)\ (aq)$	0.0106 s^{-1}	van Loon et al., 2000
$Hg(II)\ (aq) + HO_2\ (aq) \longrightarrow Hg(0)\ (aq)$	1.7 x 10^4 M^{-1} s^{-1}	Pehkonen and Lin, 1998

$Hg(0)\ (aq) + HOCl\ (aq) \longrightarrow Hg^{2+}$	$2.09 \times 10^6\ M^{-1}\ s^{-1}$	Lin and Pehkonen, 1998
$Hg(0)\ (aq) + OCl^- \longrightarrow Hg^{2+}$	$1.99 \times 10^6\ M^{-1}\ s^{-1}$	Lin and Pehkonen, 1998

Hg(II) refers to divalent Hg species

[a] The parameters are for temperatures in the range of 20 to 25°C, see references for exact temperature; temperature dependence is included in the model for the Henry's law parameter of O_3, HCl, SO_2 (Liu et al., 1997; Seigneur and Saxena, 1988), and Cl_2 (Lin and Pehkonen, 1998), and for the kinetic rate parameter of the $HgSO_3$ reaction.

[b] This reaction between HSO_3^- and H_2O_2 is fast and is treated as an instantaneous titration between SO_2 and H_2O_2. Since H_2SO_4 is a strong acid, the effect of this reaction on pH is taken into account.

3.0 EMISSIONS INVENTORY DEVELOPMENT ASSESSMENT

Emission inventories are a fundamental building block to the assessment of air quality modeling and deposition analyses and interpretation. The process of surveying, obtaining, processing, and reporting of emissions inventory activity data, emissions factors, and resulting pollutant release rates is an exceptionally resource-intensive activity. In order to simulate a 2002 annual ozone/PM/mercury episode over a domain encompassing the continental United States and a third of Mexico and Canada, significant effort was needed to prepare the model-ready inventories of mercury and the host of criteria air pollutants necessary to support this modeling.

Construction of these emissions inventories are an essential component of the WDNR's mercury modeling system. The quantitative and qualitative elements of these datasets must be verified and validated for the subsequent HgCAMx modeling to have maximum reliability and usefulness in a regulatory context. Emission estimates, source category coverage, and the temporal, spatial, and speciated nature of mercury and ozone/PM precursor emissions directly affects the reliability of the atmospheric modeling exercise. Expectations that the HgCAMx modeling system will accurately reproduce observed concentrations of gaseous, divalent, and particulate mercury as well as the other gas-phase and aerosol precursor species, can only be achieved if the emissions inputs to the modeling system closely resemble the actual mass emissions rates and chemical speciation that occurred during the period being modeled. This chapter reviews the efforts put forth to generate these emission data sets for the 2002 HgCAMx modeling and identifies key strengths and weaknesses of the inventorying exercise.

3.1 Emission Data Sources

A major source of information we relied upon in assessing the development of the emission inventories used in the HgCAMx development and evaluation was the summary document prepared by WDNR (Hetherington, 2003). This document was well organized, clear, concise, and detail-oriented in the way it presents the facts and summaries involved with the development of the modeling inventories. The document made it straightforward for Alpine reviewers to verify and validate the sources of information utilized in this exercise.

One immediate concern with the emission inventories utilized for this analysis was the appropriateness of the emissions for the year of analysis. As indicated in the conclusions section of the HgCAMx report (Yarwood et al., 2003a), "An annual 2002 modeling database was developed to test and evaluate the CAMx mercury model." In reality, a 1999 calendar year inventory of mercury and criteria emissions was developed and used to simulate the 2002 year. The effects of known and implemented control programs (e.g., Title IV) could have a significant impact on coal-burning EGUs, a major contributor to mercury emissions. For this reason, the WDNR should consider performing appropriate sensitivity analyses and emission comparisons once these actual 2002 data become available.

3.1.1 Mercury Emissions Inventory

As part of the HgCAMx mercury model development program for Wisconsin and the Great Lakes Region, the WDNR developed an annual 1999 emissions inventory for mercury based on the 1999 National Emissions Inventory (NEI) version 3 final, Electric Power Research Institute (EPRI) data for U.S. EGUs, Great Lakes State data from 1999, and Canadian emission data sources. Most of these sources of emissions were at the time, and are now during this review, the best available mercury

emission data sets. Recent modeling conducted at EPA for purposes of developing regulatory modeling support of mercury emission reductions under the Clear Skies Act (EPA, 2003a) utilized the 1999 NEI and EPRI data in the same fashion as this mercury study and analysis. Addtionally, the supplemental effort put forward by WDNR staff to augment these data with regional, local, and source specific emissions is commended. Utilizing Great Lakes State data, not only for those sources that have the greatest impact on mercury transport and deposition, but all sources of mercury emissions, enhanced the quality of the emission estimates. To date, few additional mercury studies have been conducted for domains which intersect with that of this analysis. For those that have, similar, if not identical starting data sets of U.S. mercury emissions were used.

A review of the annual mercury emission totals between the final ENVIRON (Yarwood et al., 2003a) and WDNR summary documents indicate a discrepancy between the total annual emissions reported by ENVIRON and those totals reported by WDNR. This discrepancy has been identified as the difference between EPA's 1999 NEI versions 3 final and draft. ENVIRON's modeling inventories were based on the draft release of these inventories, while WDNR's summaries are based on the more recent final version. Referring to Table 1 from the WDNR summary and comparing with the totals in ENVIRON's Table 3, we have summarized the emissions total differences between the two groups in Table 3-1. As the modeling totals are 15% higher than the WDNR reported annual emissions, this discrepancy should be corrected prior to any future modeling efforts of the 2002 annual period.

3.1.2 Ozone and PM Precursor Emissions Inventory

The emissions inventory all non-mercury species was based on the Midwest Regional Planning Organization (MRPO) "Base D" emission inventories. In turn, these data were largely based on EPA's 1999 National Emission Inventory (NEI) version 2.0 with several corrections made by the MRPO. This "Base D" includes the following improvements:

> **Point Sources.** Utility temporal profiles were based on analysis of CEM data;

> **Area Sources.** Ammonia emissions were based on new version of the Carnegie Mellon University (CMU) model;

> **Mobile Sources.** Emission estimates were based on MOBILE6; and

> **Other.** New Canadian emissions inventory and dust emissions were reduced by 99%.

This inventory, like most produced and modeled with the help and guidance of the MRPO, underwent extensive QA/QC by MRPO staff and stakeholders. The quality control and quality assurance steps taken with these data are documented (Janssen, 2003) and were examined for this review. However, like most emission inventories, the MRPO has more recently released a revised "Base E" ozone and particulate matter precursor emission inventory update. This inventory reflects the following emission revisions from their previous "Base D" (MRPO, 2003a):

> **Mobile Sources.** Corrected diesel emissions ("Base D" did not include most of these emissions);

> **Ammonia.** Monthly and hourly livestock emissions are based on new temporal profiles from Rob Pinder. Additionally, dairy cow emissions are based on Pinder's model. The monthly

fertilizer application emissions are derived using a consistent national profile and emissions were eliminated for people and pets (dogs and cats);

> **Dust.** Emissions reduced to reflect the transportable fraction of fugitive dust;

> **Fires.** Eliminated NEI and CMU fire emissions; and

> **Biogenics.** Used BIOME3 with updated meteorology and PAR values.

Although reviewed reports indicate that "Base D" was used for the mercury modeling assessment, conversions held with WDNR modeling staff suggest that newer, revised modeling has taken place with "Base E." These revised modeling data do not appear to be included in the ENVIRON final analysis and may not have been completed at the time of the study.

3.1.3 International Inventories

Similar to most recent regional- or continental-scale studies conducted in the last few years, the available emissions information from Mexican and Canadian sources was limited and for those data that were available, confidential. To the credit of the WDNR inventory development team, the province of Ontario was contacted directly and the usage of some confidential 1999 point sources was granted. These point source data were consolidated with the area source emissions by Ontario and provided to WDNR as an area source file. For the remainder of the Canadian domain, incomplete 1995 emission inventories were utilized and did not present as complete a picture as required for a comprehensive analysis. Mexican emissions were provided by the North American Council for Environmental Cooperation (CEC) and were for only a limited number of point sources. The area and mobile source data available at the time of the study was not in a form that would lend itself to modeling and was therefore excluded from the study. Recent advancements in the collection and reporting of emission inventories in Mexico and Canada may soon lend themselves to better, more consistent inventories with those currently available in the United States. When those sources of data become available, it would be worthwhile for the WDNR to explore their added value.

3.2 Revisions and Augmentations to Emission Inventories

Rarely is an inventory developed for national applications (e.g., 1999 NEI) complete, comprehensive, and representative of every gridded domain modeled within that inventory's national coverage. For this reason, emission inventories need to be modified and augmented with additional data from sources more local to the modeling domain. In addition to converting the formats into files that can be read by the emissions processors, a list of quality assurance routines to try and catch any missing or "out-of-range" parameters which may cause issues in the processor runs is completed. Some of these routines will result in modifications to the data. WDNR has provided in its emission inventory preparation documentation the identification, QA/QC, augmentation and rationale for the changes required in these data sets for purposes of the mercury modeling assessment. In fact, the procedures cited in the revisions to these inventories are commonly utilized and encouraged by the emissions modeling community.

3.2.1 Emission Inventory Augmentation

Mercury emission inventories developed for the Great Lake States (GLS) were consolidated with emissions from the base 1999 NEI. These point, area, and mobile source data were previously

determined to have been more representative and comprehensive in their coverage and magnitude for the area of greatest impact to this study.

> **Point Sources.** Facility and unit matches were conducted during this augmentation step with GLS data taking precedence over the NEI generated emissions. In cases where matches to facilities or units could not be established, both sets of information were kept. Additional cited improvements to the point source mercury emissions include the calculation of previously unreported mercury emissions at combustion sources in Wisconsin using EPA approved emission factors and corrections to mercury emissions at multiple facilities using recommendations from the Mercury Advisory Group of WDNR.

> **Area Sources.** Although the area source augmentation and replacement steps occurred at a State-wide level instead of county by county, emissions of this nature are typically estimated first at the state level and then allocated to county based on ratios of activity or spatial surrogate data (e.g., population). This process is representative of other replacement activities for area source emission categories.

> **Mobile Sources.** Mobile sources not taken from the 1999 NEI or its derivatives were generated using similar means (e.g., MOBILE model) and replaced using approved methods. The only improvement opportunity encountered in the development of local mobile source emissions was the usage of MOBILE5 for the GLS replacement inventories. Although not documented, it is presumed that the revisions made to account for localized conditions include improvements to VMT, vehicle mix, vehicle speeds, etc. which modeled in MOBILE6, provide a more accurate representation of the onroad mobile source emissions.

3.2.2 Quality Assurance/Quality Control (QA/QC)

The QA/QC completed by WDNR during the development and preparation of the emission inventories for this analysis is well documented and comprehensive. The procedures used to QA/QC these data follow the outline of EPA and MRPO identified functions (EPA, 2003; MRPO, 2003b) and are designed to correct anomalous data values.

> **File Conversion.** For all source categories within the modeling domain, procedures were developed and data were verified prior to and after the conversion from the inventory's native format to NEI Input Format (NIF) to be read by the Emission Modeling System (EMS-2003). These procedures verified data integrity and relational data structure. Duplicate records generating potentially double counted emissions were dropped from the tables.

> **Missing Data.** WDNR reviewed area and point source emission inventories in Wisconsin, Minnesota, Illinois and Michigan to determine if sources were missing or non-valid. In cases where these occurrences were located, these sources were investigated and appropriate replacements were made.

> **Out of Range Values.** During the QA process, WDNR applied standard deviation checks to identify and correct out-of-range emission and physical stack characteristic values. Where non-valid data were identified, values were modified or emissions were removed consistent with practiced and documented procedures.

> **Emission Estimation.** Although typical during the inventory development process, WDNR performed the additional step during the conversion of mass inventories to modeling inventories of verifying emission estimation calculations using fuel throughput and emission factor data. Where these two data elements were available via the NEI or GLS inventories, final emission calculations were verified with these factors and associated control parameters.

3.3 Source Category Coverage

Another important aspect of emission inventory development is the coverage of source categories which emit the pollutants of interest. In this case, extended efforts were taken by WDNR to identify and quality assure emissions from source categories typically associated as mercury emitting. A four step process was developed and implemented by WDNR in the development of the mercury emission inventories and is presented with comment below.

> **Identify Mercury Emitting Sources.** Using published data and emission factor information, WDNR identified source categories consistent with emissions of mercury. Comparing these lists with summaries generated from the WDNR inventories provided an overview of source categories within the modeling domain with the potential to emit mercury. In cases where sources were identified with the potential to emit, but positive emissions were not found, sources were flagged.

> **Calculate mercury emissions from identified sources.** From the previous identification step, once a source was found to be a potentially emitting mercury source, emissions were calculated using available emission factor and fuel throughput values. Instances occurred where non-mercury emissions were identified for a source or source category and mercury emissions were listed as zero. In these cases, and where identified as a potentially mercury emitting category, WDNR used published emission factor data to generate a consistent mercury value to the other reported pollutants. If mercury emissions were already identified at a source, these emission calculations were verified using the methods described in the previous section of this report.

> **Identify sources with limited emission estimation data.** Following the comparison of existing mercury emitting sources, WDNR reviewed lists of categories which contained partial information necessary to generate mercury emission estimates. Further review was conducted at WDNR to augment these missing data and where enough information was discovered to supplement the original data, mercury emissions were calculated and integrated into the modeling inventory.

> **Identify missing sources.** Finally, following the steps presented above, WDNR identified an additional source (forest fires in Wisconsin) within the modeling that was not represented in the local (e.g., Great Lakes) domain and attempted to estimate (domain-wide) mercury emissions from this category. However, due to the relatively small magnitude of these emissions and absent a reasonable method for spatially allocating these emissions, these data were not included in the final modeled inventory.

3.4 Use of Emissions Data

The development of emissions inventory information and the processing of these data into air quality model-ready format must be carefully matched to the nature of the modeling to be performed and the expectations one has for desired model representativeness, accuracy, and precision. Inventory development must also be cognizant of and strive to quantify the limitations of the data sets. In the present initial application of HgCAMx to the annual 2002 period, it seems to make sense to utilize the annual inventories developed by WDNR as a first step in attempting to develop seasonal, monthly and diurnal variations in emissions rates and the spatial distribution of emission sources and categories.

Annual emissions were generated by emissions modelers at the MRPO and WDNR as described in the preceding sections. However, it appears from the ENVIRON final report that only three representative days (weekday, Saturday and Sunday) from two months (January and July) from two episodes (January 2000 and July 2001) were actually used to represent an *entire* year's anthropogenic mercury, ozone, and particulate matter emissions for 2002. If correct, then this statement in the report contradicts the text in the conclusions section of the ENVIRON report which states, "(m)odeling an entire year is important to capture seasonal cycles in mercury deposition." Apparently, these three days from these two months were used to generate the full set of 365 modeling days needed to simulate the annual episode. Because the ozone and PM precursor inventories were already available in EMS processed form for the weekday/Sat/Sun January and July periods, we can only deduce that mercury emissions were processed in a similar fashion to generate similar data sets.

Thus, it appears that a significant shortcut was taken in constructing the daily emissions information supplied to the HgCAMx model, in spite of the fact that VOC, NOx, and PM inventories are available for the region with more realistic temporal and month-to-month variation that that accorded by "three days from two months of the year". We believe that at a minimum representative monthly emissions of mercury should have been generated to account for the temporal nature of the largest contributing source types, especially since the focus of the modeling was on mercury. Given that one purpose of the HgCAMx modeling was to evaluate the model's ability to simulate the annual impact of mercury emissions from various sources in the region throughout the study year, we find that the model-ready inventories apparently supplied to HgCAMx in this exercise introduce potentially serious limitations to the adequacy of the modeling. In short, the inventories developed by WDNR appear to have been inappropriately utilized in constructing the actual model-ready inputs to HgCAMx for the annual modeling.

3.5 Synthesis of Emissions Inventory Development

3.5.1 Emission Data Sources

Our review of the emission data sources used in the development of an annual mercury emissions inventory for the modeling domain of this analysis suggests that these inventories of mercury were, and still are, among the most recent and representative available given today's state-of knowledge. Additionally, extended efforts were taken to secure Canadian emissions from the province of Ontario that typically would be confidential in nature. These additional sources and their use in this analysis increased the credibility of the results. In our view, the 1999 emissions inventory of mercury emissions has been developed in a credible fashion and in some cases, has exceeded the quality of federally managed data.

However, there are some fundamental concerns with the model-ready emissions data sets apparently supplied to HgCAMx in this study. First, a reconciliation of the WDNR generated emissions and those modeled by ENVIRON needs to be conducted. There is a 15% difference in the total national mercury mass between the summaries presented in summary reports. Should emissions have been modified between the two steps, these modifications needs to be documented. Secondly, there is an inconsistent correspondence of the base emissions data to the episode of interest. No efforts were expended to generate an actual inventory for the 2002 calendar year. Since the ambient monitored data were from the calendar year 2002, it would make the most sense to compare emissions from 2002 to validate model results. The effects of known and implemented regulatory programs could have a significant impact on major contributors to mercury emissions. Finally, recent advancements in Mexican and Canadian emission development and inventory distribution exercises have provided additional data for North American air modeling analyses. When available, these inventories and most recent U.S. data should be processed and compared to existing results.

3.5.2 Revisions and Augmentations to Emission Inventories

Our review of the revisions and augmentation steps applied to strengthen the validity of the emission inventories suggest that these data are a credible representation of the period for which they were generated. In fact, the methods applied by WDNR to develop these inventories and make them regionally-representative are commended. We found no weaknesses with the procedures used to prepare these data for modeling.

3.5.3 Source Category Coverage

Our review of the source categories reported in the emission inventories developed by WDNR suggest that these data are completed and comprehensive in their coverage and magnitude. We additionally recognize the extended efforts put forth by WDNR to locate, identify, and estimate alternate, unreported categories and sources of mercury emissions. We found no weaknesses with the methods and coverage of sources in the inventories prepared for modeling.

3.5.4 Use of Emissions Data

Our review of the usage of the mass emissions inventory data for this analysis suggests that the application of limited temporal variation to prepare CAMx input files was inappropriate for the modeling of an annual 2002 episode. In fact, the assumptions used to generate model-ready inputs for HgCAMx potentially undermine the utility of the present model performance evaluation. Contrary to the conclusion section of the ENVIRON final report, an annual emissions inventory was not used to capture the seasonal cycles in mercury deposition. In fact, only three ‘representative’ days from two seasons were modeled, with the remaining modeling days constructed using an unclear interpolation procedure. Thus, we believe that the actual model-ready emissions data sets input to HgCAMx may well have been inadequately representative of the collection of actual days making up the annual episode, thereby incompletely capturing the seasonal cycles of the major contributing source categories. The reliability of the HgCAMx model verification results are thus open to question.

Table 3-1. Comparison of WDNR and ENVIRON Reported Hg Annual Emissions.

	WDNR Table 1	ENVIRON Table 3-3	Difference
All US Hg Point	109.74	130.03	20.29
All US Hg Onroad	20.36	20.36	0
All US Hg Area	9.54	15.869	6.329
All US Hg Off-Road	7.28	7.28	0
Total US Hg	146.92	173.544	26.624

4.0 EMISSIONS MODELING ASSESSMENT

The emissions modeling peer-review process consisted of three basic operations customarily scrutinized during such a review. These included: (a) the temporal distribution of emissions data to the hourly requirements of the modeling episode, (b) the spatial allocation of emissions sources to the gridded modeling domain, and (c) the disaggregation of the mercury emissions estimates into the species categories required by the atmospheric model. To evaluate the emissions modeling performed on the mercury inventory, AG has reviewed both the inventory documentation prepared by WDNR, and the modeling documentation prepared by ENVIRON.

WDNR provided a document summarizing the preparation of the emissions inventory (Hetherington, 2003). This document included the sources for all portions of the inventory, summaries of the mercury emissions data and an overview of the QA/QC and processing steps taken during the emissions modeling activities. Our independent review of these activities confirmed that the WDNR emissions modeling methodology conformed to the standard EMS-2003 processing rules, and that updates or alterations to the inventory data are documented. The ENVIRON final model report also included a section on the development of the emissions inventories and we reviewed this document also for consistency in the emissions modeling approach.

To further evaluate the major emissions modeling components, we received from WDNR the pre-merged model ready ASCII files produced by EMS-2003, along with the EMS-2003 run summaries and reports. These were evaluated against the lookup and cross-reference tables available in the EMS-2003 release in conjunction with updates to the standard lookup tables provide by WDNR. In parallel, we obtained the merged model ready files from ENVIRON and utilized these files for overall comparisons. These files were reviewed using Alpine's in-house emissions modeling QA software (developed for typical EMS-2003 and SMOKE applications) to spot-check the adequacy of the profiles applied to the mercury emissions and to evaluate whether the level of detail included in the distributions were adequate.

The HgCAMx emissions input files for the non-mercury species were produced using the Midwest RPO "Base D" emissions data sets and EMS2002 emissions modeling configuration. Although The Base D inventory utilized by ENVIRON was the most current emissions inventory available at the time, there are several known shortcomings in the "Base D" emissions configuration that will affect any modeling based on this inventory. The most significant differences between the "Base D" and the subsequent "Base E" emissions versions that potentially affect mercury emissions modeling are (Janssen, 2003):

> **Updated and Improved Temporal Profiles for "Base E".** The point, other area, nonroad, and mobile source temporal profiles used in the "Base D" version are predominately "flat" with little or no hourly variability. These profiles have been improved for "Base E"; and

> **Improved and Corrected Spatial Allocations.** The spatial allocation factors in the "Base D" versions of the inventory were, in some cases, mis-assigned. The most significant problem was the assignment of farmlands to the default profiles. The spatial surrogates for other area, nonroad and mobile source have been updated for "Base E".

Any additional modeling should be based on Base E, or the most current inventory revision available.

Subsequent review of the mercury portion of the emissions inventory focused on the correction of temporal and spatial allocation of the mercury species.

4.1 Temporal Distribution Review

The review of the temporal allocation consisted of three steps:

> **Review of EMS-2003 Processing Logs and Reports Provided by WDNR**. EMS-2003 prints summary reports before and after the application of temporal profiles, a warning list of emissions that are dropped because of incorrect temporal profiles, and a listing of sources which receive "default" profiles due to lack of profile information. The review of these reports did not reveal any systematic problems with the mercury portions of the inventory.

> **Review of Temporal Profiles Applied to Mercury Source Categories.** The major SCC contributors for each source category were determined by reviewing the WDNR Emission Inventory documentation. The SCC codes accounting for 75-80% of the emissions in each source category were selected and checked for temporal profile assignment. The results of this comparison are presented in Table 4-1. This comparison did not reveal any mercury source categories being assigned to incorrect temporal profiles. The profiles used are characteristic of those used for photochemical modeling. However, the profiles are very general and constitute an area where potential refinements can be made in future mercury modeling inventories.

> **Review of Hourly Emissions Distributions**. The hourly emission distributions were plotted for each episode day and mercury species. These plots were reviewed to assure that temporal distributions were appropriate for each source type. Figures 4-1 through 4-5 show examples of hourly emission distributions for one episode day and one specie. This review indicated that temporal resolution typical of the SCC emissions classes was consistently evident in all of the source categories.

4.2 Spatial Distribution Review

The spatial allocation process was reviewed in the following steps:

> **Review of the EMS-2003 Spatial Processing Logs and Summaries**. Summary reports for the spatial allocation of area, nonroad, and on-road categories were not available for review.

> **Review of Spatial Surrogate Assignments**. The EMS-2003 cross-reference files were examined to determine that the major SCC categories were being assigned to appropriate spatial distributions. The profile categories for each of the area SCCs are presented Table 4-3.

> **Review of Spatial Allocation Plots**. The spatial distributions of emissions were plotted by hour for each episode day and mercury species. These plots were reviewed to assure that spatial distributions were characteristic for each source type. Figures 4-6 through 4-20 provide examples of one episode day and hour for each of the three Hg species.

Our review indicated that adequate spatial distributions were being displayed in all of the source categories. The area and nonroad plots show clear evidence of both population centers and agricultural areas, while the on-road motor vehicle concentrates emissions into population centers that reflect the highway distributions.

4.3 Speciation Allocation Review

The standard EMS-2003 speciation profiles were updated to include mercury species for the major contribution SCC codes, and to provide a default profiles for any Hg species. The speciation application was evaluated in two phases:

4.3.1 Review of Major SCCs to Determine Assignment of Speciation Profiles.

The list of major SCC codes (Table 4-1) was compared to the speciation profile and definition datasets (profile.sas7bdat and spprcr.sas7bdat) to determine the assignment of Hg profiles used in the speciation application. For all of the major contributor SCC codes in the point, non-road and on road categories, an SCC specific speciation profile is assigned. In the area source category, although VOC profiles were available, Hg profiles were not specifically allocated for the following categories:

> 2801000 – Miscellaneous Area Source, Agricultural production, Crops;

> 2311010 – Industrial Process, Construction, General Building; and

> 2311030 – Industrial Process, Construction, Road Construction

The default speciation profile was applied to these categories. This profile may be adequate, considering the general nature of these area source categories. However, they should be reviewed in any future Hg inventory development activity.

4.3.2 Verification of Emissions Totals Before and After Application of Profiles.

To verify that the emission speciation profiles were being applied correctly, the Hg emissions summaries before and after speciation were compared to assure that Hg mass was not being lost. The speciation profiles for mercury are mass-conserving, so the pre- and post-speciation totals should be equal. Calculations were preformed on representative days from the winter and summer episodes, and the mass totals were found to be consistent.

4.4 Synthesis of Emissions Modeling Review

Overall, the emissions modeling review indicates that the mercury portion of the emissions inventory has been modeled with due care. We recommend the WDNR consider implementing the following updates to the emissions inventory modeling process in subsequent applications of HgCAMx:

> Update the non-mercury emissions inventory to utilize the “Base E” inventory rather than to outdated “Base D” inventory; and

> Review of default application of the speciation profiles to determine if the default is appropriate.

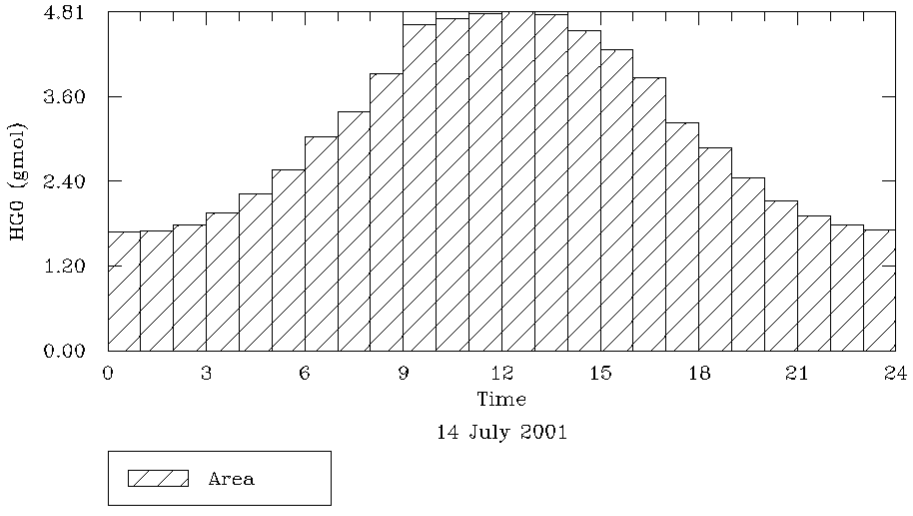
Table 4-1. Temporal Profile Assignments for the Major Contributing Hg Sources by Emissions Category (U.S. Only).

Major SCC	Description	Emissions (Tons/yr)	Profile Definition*
Area Sources			
2801000	Misc.Area, Agricultural Crops	1.282	26
2311010	Industrial Process, General Building Const.	1.278	26
2104004	Stationary Source Fuel – Residential Distillate Oil	1.154	26
2861000	Misc Area, Fluorescent lamp breakage	0.997	24
2311030	Industrial Process, Road Construction	0.971	26
315030	Industrial Process, Laboratries	0.836	24
315025	Industrial Process, Dental Alloy Production	0.696	24
NonRoad Sources			
2270002	Off-Highway Diesel Construction and Mining	2.315	26
2270005	Off-Highway Diesel Agricultural Equipment	2.227	26
2285002	Railroad Equipment, Diesel	0.708	24
2280002	Marine Vessels, Commercial Diesel	0.589	24
Motor Vehicles			
2230074	Highway Vehicle –Diesel HDDV 8A,8B	11.314	2001-13
2230073	Highway Vehicle – Diesel HDDV 6,7	3.112	2001-13
2230071	Highway Vehicle – Diesel HDDV 2B	1.842	2001-13
2201001	Highway Vehicle – Gasoline LDGV	1.337	26
2230072	Highway Vehicle – Diesel HDDV 3,4,5	1.314	2001-13
Point Sources			
101002	Ext Comb Boilers, Electric Gen. Bituminous Coal	47.194	33
303013	Industrial Process, Primary Metal Production, Gold	10.593	24
399999	Industrial Process, Misc Processes	7.227	24
503005	Waste Disposal, Solid Waste Industrial Incineration	6.668	24
501001	Waste Disposal, Solid Waste Government Incineration	5.742	24
101003	Ext Comb Boilers, Electric Gen. Lignite	4.422	33
502005	Waste Disposal, Solid Waste, Commercial/Industrial Incineration	3.793	24
301008	Industrial Process, Chemical manufacture, Chloro-alkali	3.406	24

* Description of temporal profiles:
24 – 24 hour distribution with no variation
26 – variable distribution with emission peaking at 2:00 PM
33 – variable distribution with emissions peaking at 4:00 PM
2001-2013 – facility specific distribution for on-road motor vehicle sources.

Table 4-2. Spatial Profile Assignments for the Major Contributing Hg Sources by Emissions Category (U.S. Only).

Major SCC	Description	Emissions (Tons/yr)	Profile Type
Area Sources			
2801000	Misc.Area, Agricultural Crops	1.282	Agriculture
2311010	Industrial Process, General Building Const.	1.278	Population
2104004	Stationary Source Fuel – Residential Distillate Oil	1.154	Population
2861000	Misc Area, Fluorescent lamp breakage	0.997	Population
2311030	Industrial Process, Road Construction	0.971	Population
315030	Industrial Process, Laboratries	0.836	Population
315025	Industrial Process, Dental Alloy Production	0.696	Population
NonRoad Sources			
2270002	Off-Highway Diesel Construction and Mining	2.315	Population
2270005	Off-Highway Diesel Agricultural Equipment	2.227	Agricultural
2285002	Railroad Equipment, Diesel	0.708	Railroad
2280002	Marine Vessels, Commercial Diesel	0.589	Water
Motor Vehicles			
2230074	Highway Vehicle –Diesel HDDV 8A,8B	11.314	Highway
2230073	Highway Vehicle – Diesel HDDV 6,7	3.112	Highway
2230071	Highway Vehicle – Diesel HDDV 2B	1.842	Highway
2201001	Highway Vehicle – Gasoline LDGV	1.337	Highway
2230072	Highway Vehicle – Diesel HDDV 3,4,5	1.314	Highway



Gridded Emission Totals Area in the 36

Figure 4-1. Daily Emissions Distribution - Area Sources.

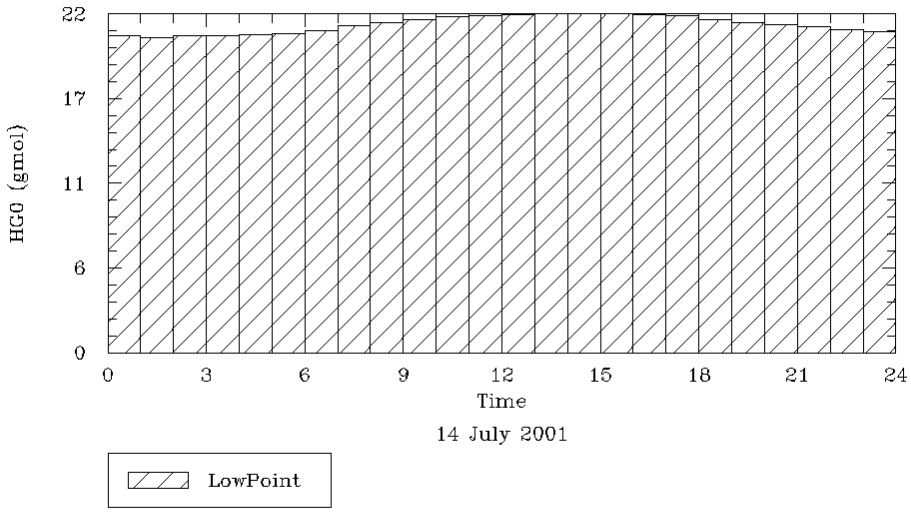


Figure 4-2. Daily Emissions Distribution - Non-Elevated Point Sources.

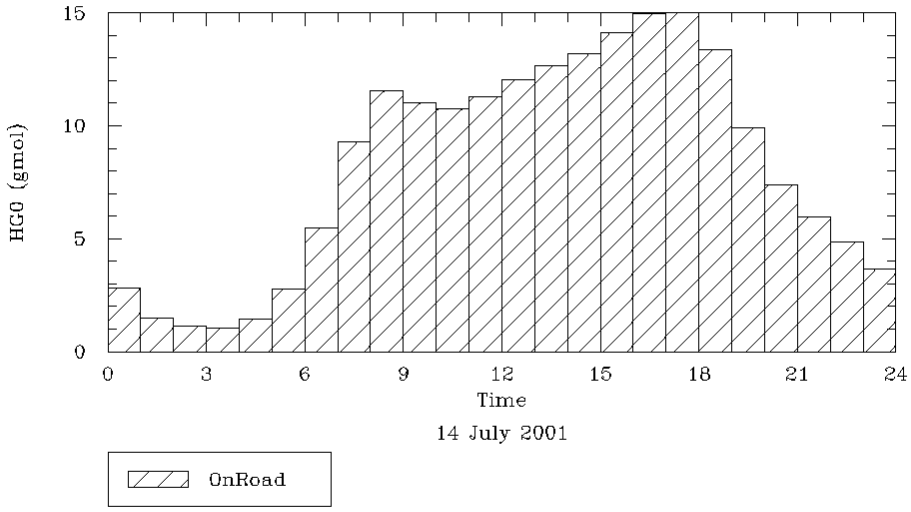


Figure 4-3. Daily Emissions Distribution - On-Road Motor Vehicles.

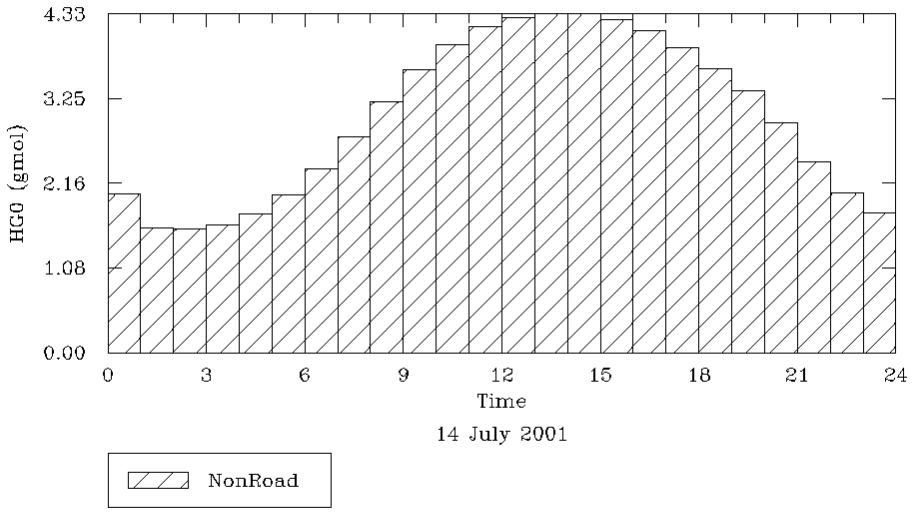


Figure 4-4. Daily Emissions Distribution - Non-Road Motor Vehicles.

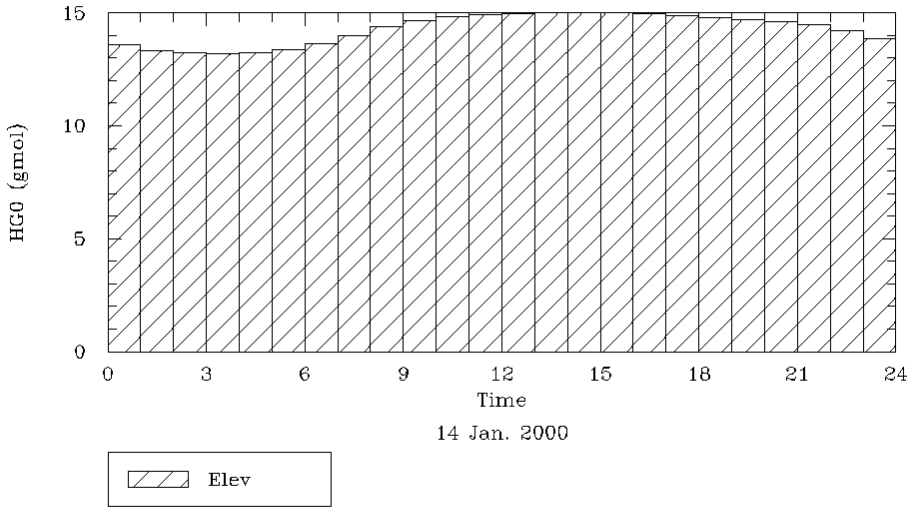


Figure 4-5. Daily Emissions Distribution - Elevated Point Sources.

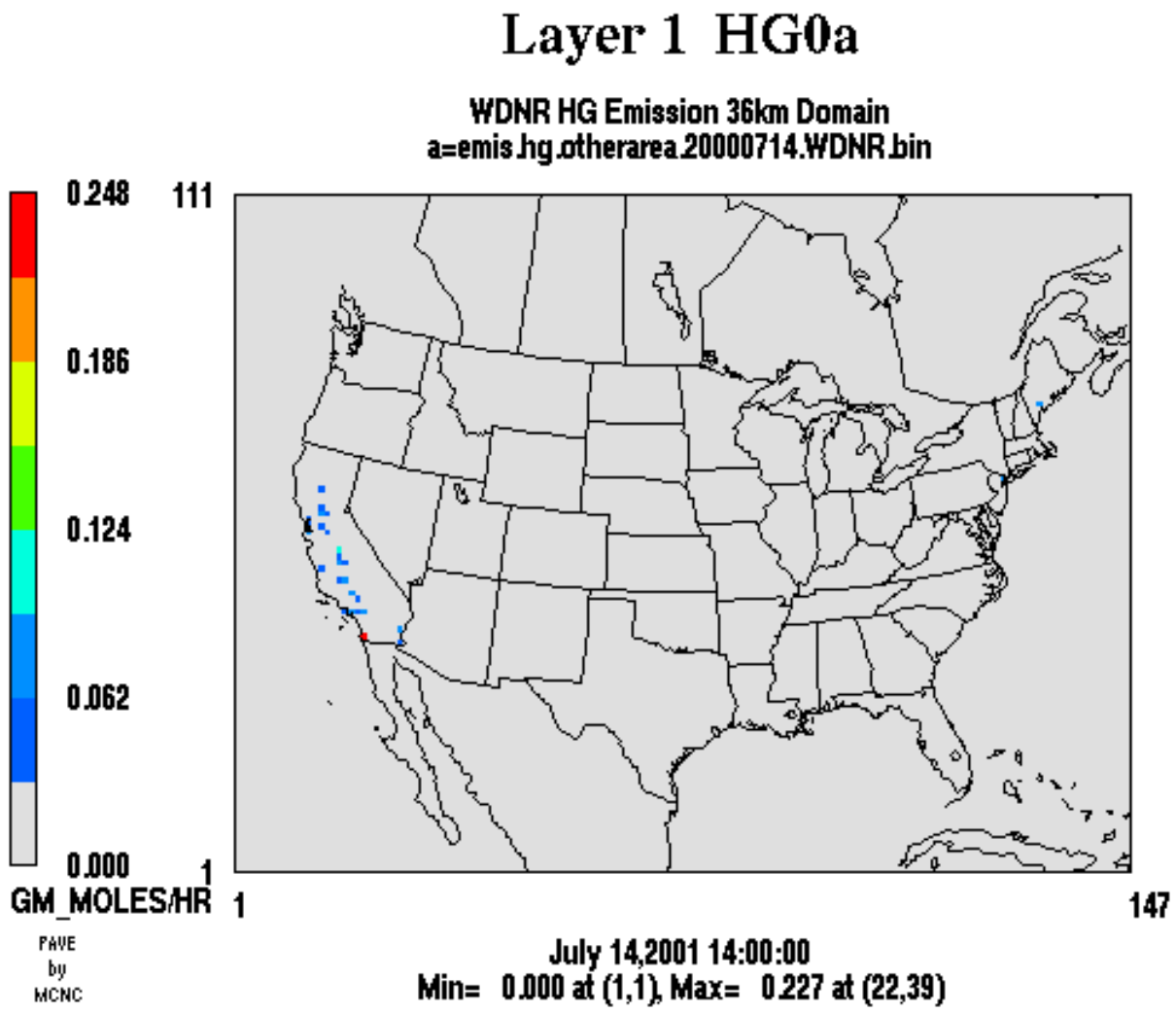


Figure 4-6. Area Source Spatial Allocation of HG0 Emissions.

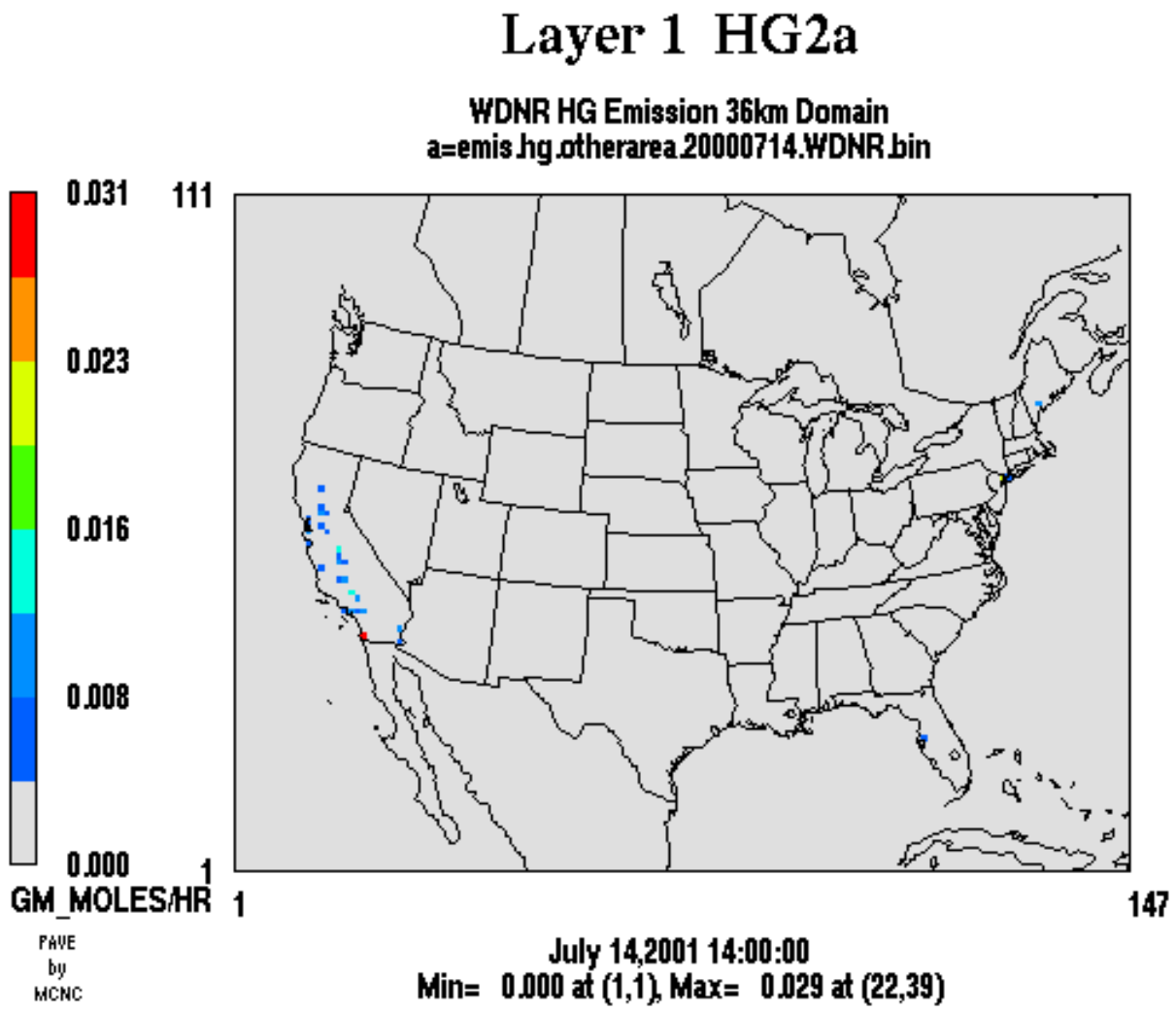


Figure 4-7. Area Source Spatial Allocation of HG2 Emissions.

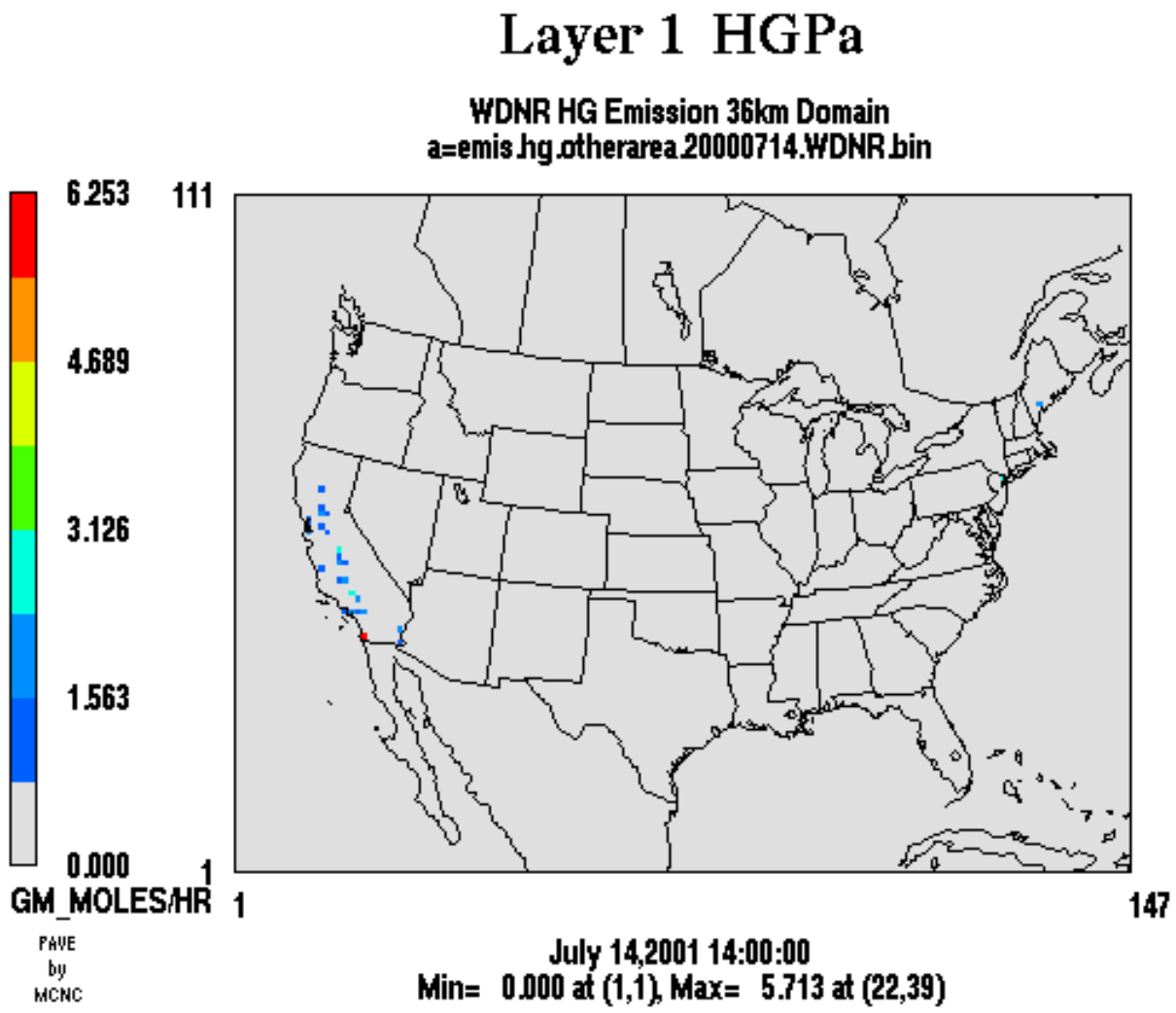


Figure 4-8. Area Source Spatial Allocation of HGP Emissions.

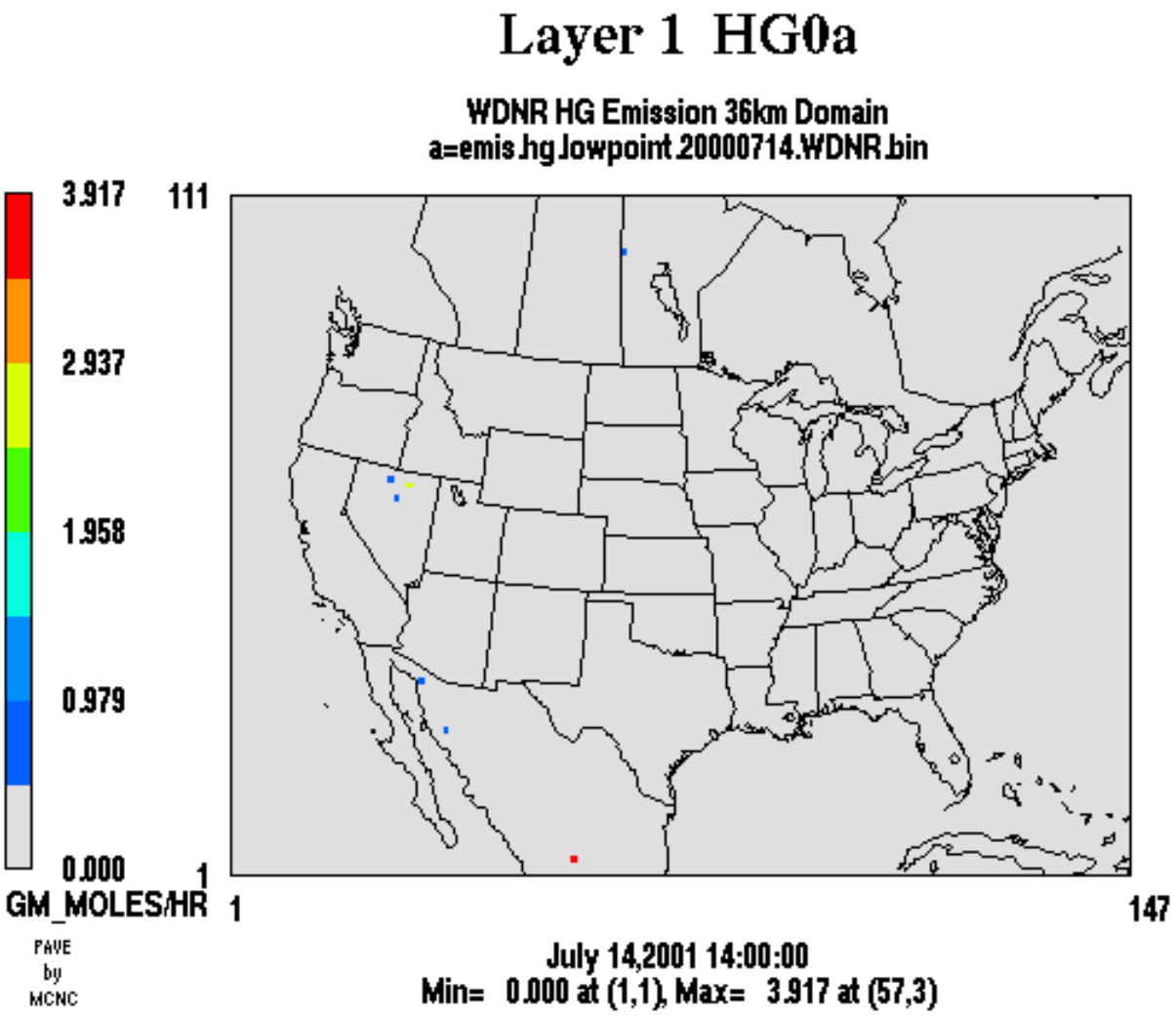


Figure 4-9. Low-Level Point Spatial Allocation of HG0 Emissions.

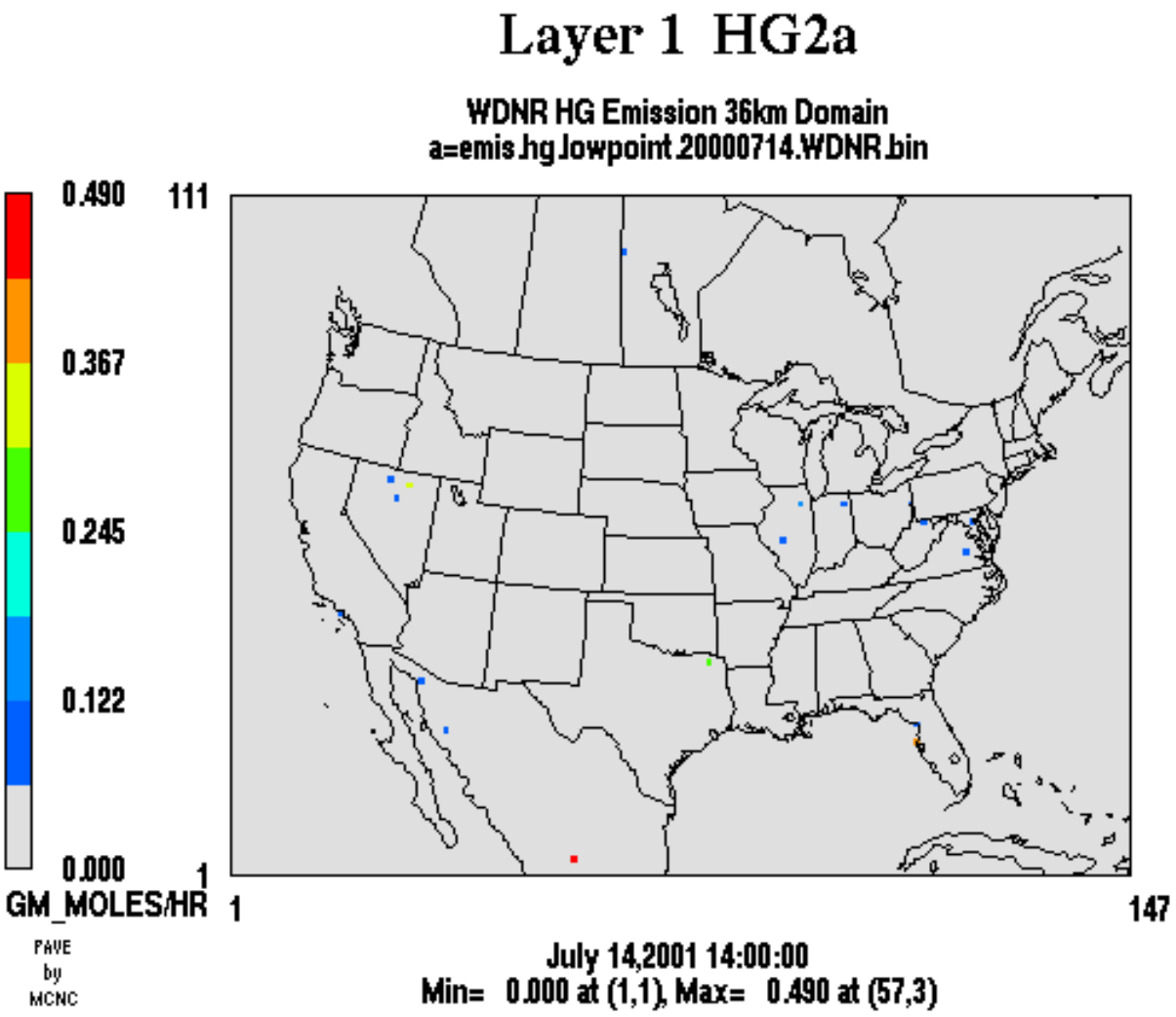


Figure 4-10. Low Level Point Spatial Allocation of HG2 Emissions.

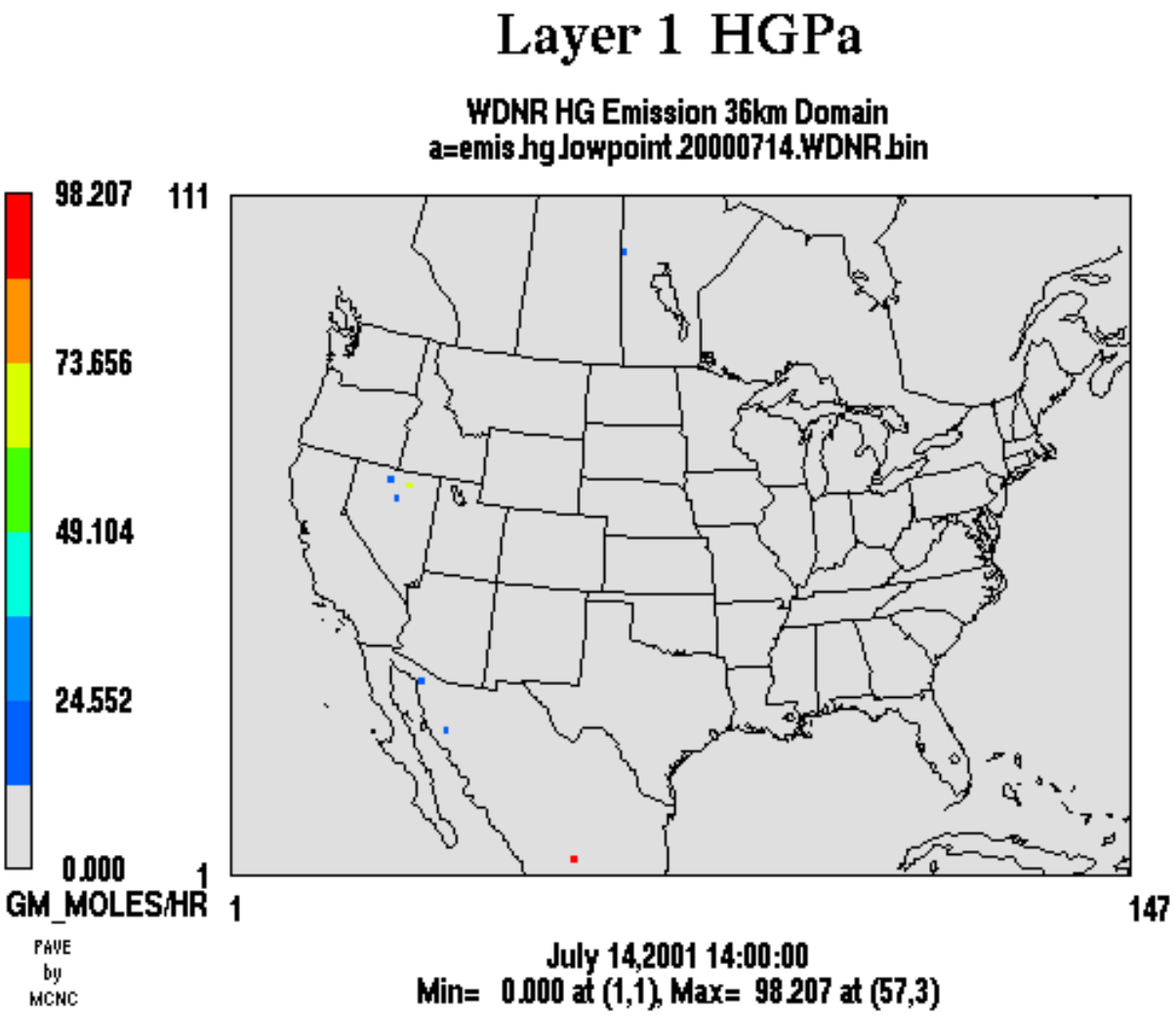


Figure 4-11. Low Level Point Spatial Allocation of HGP Emissions.

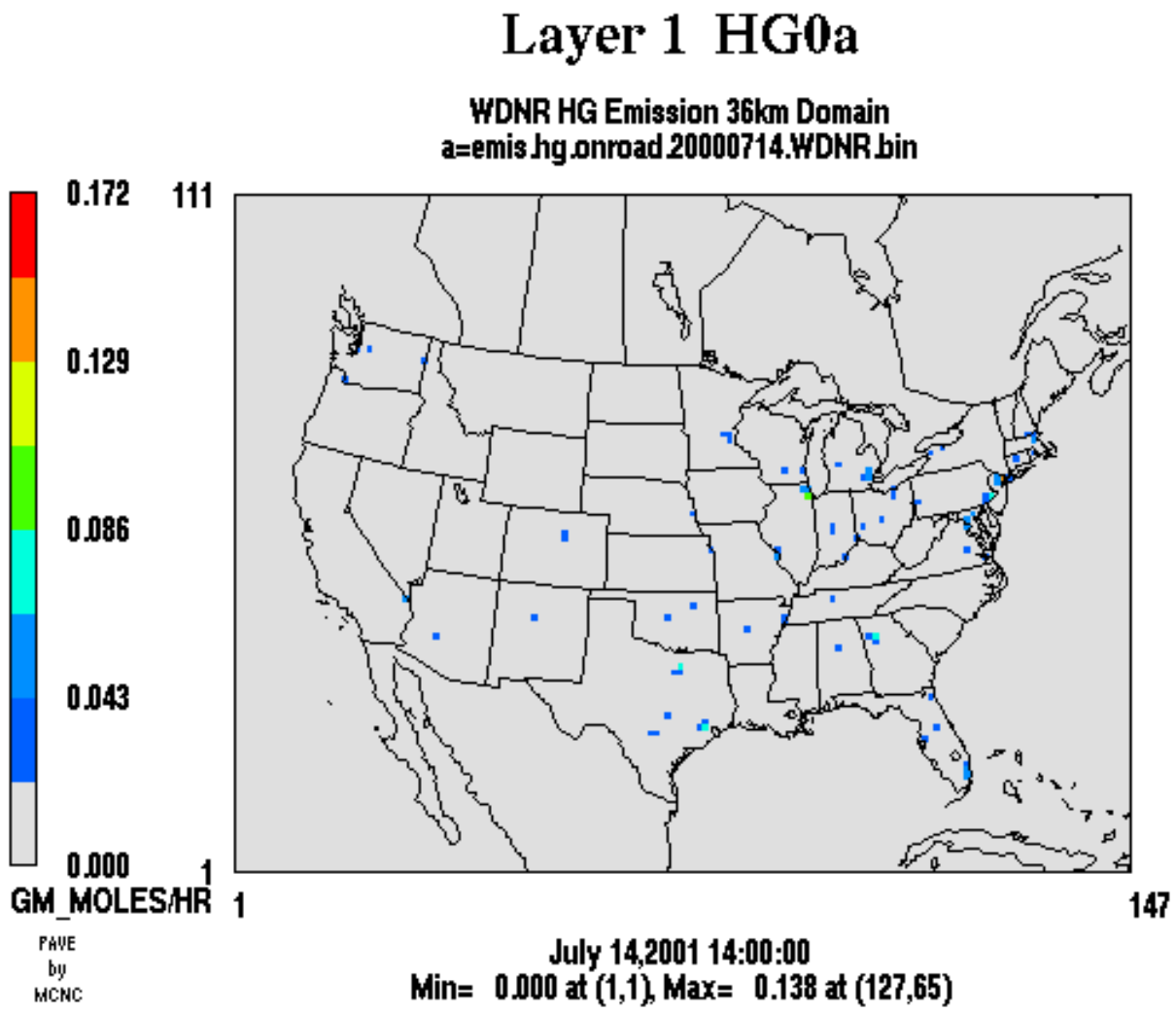


Figure 4-12. On-Road Motor Vehicle Spatial Allocation of HG0 Emissions.

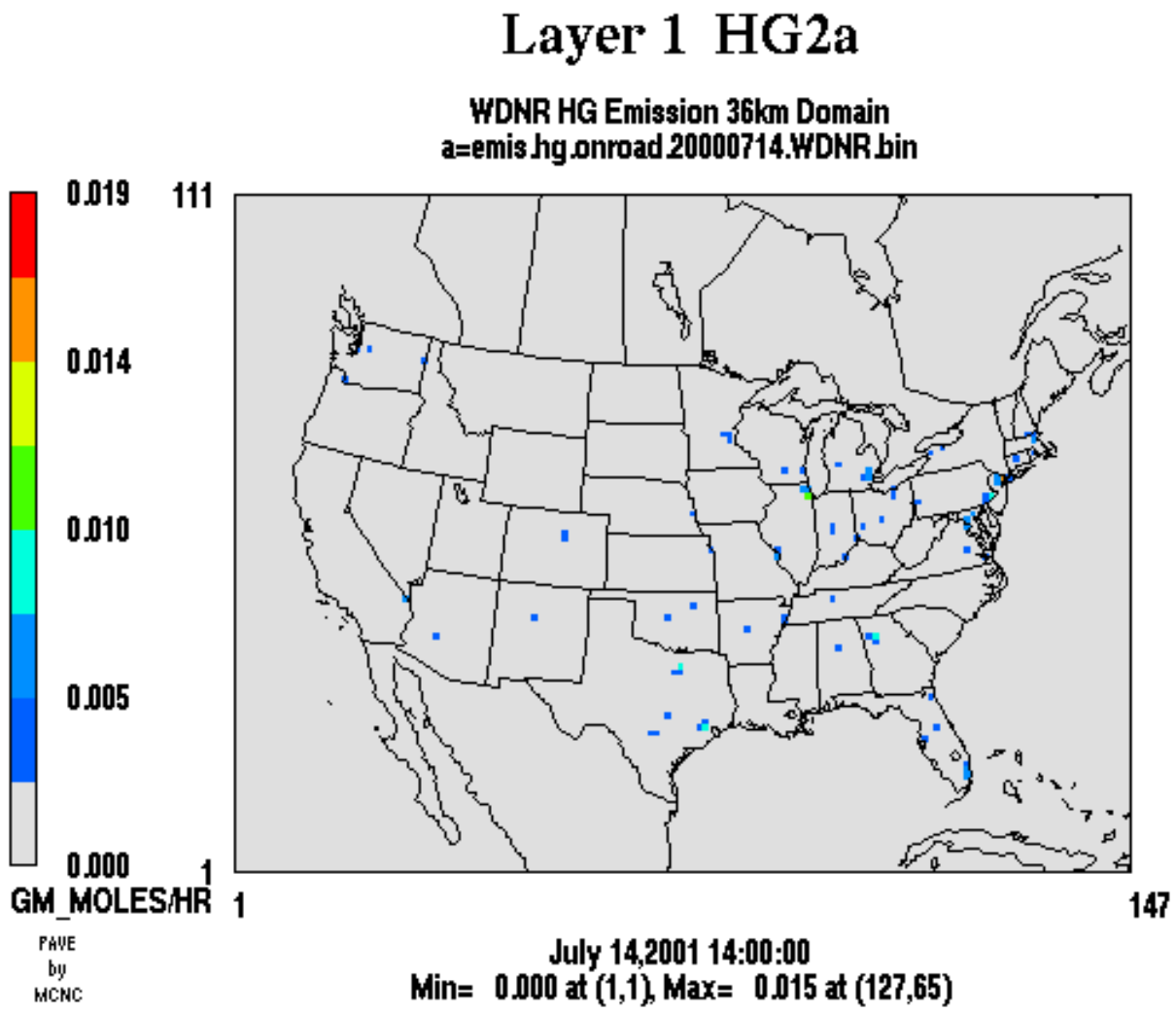


Figure 4-13. On-Road Motor Vehicle Spatial Allocation of HG2 Emissions.

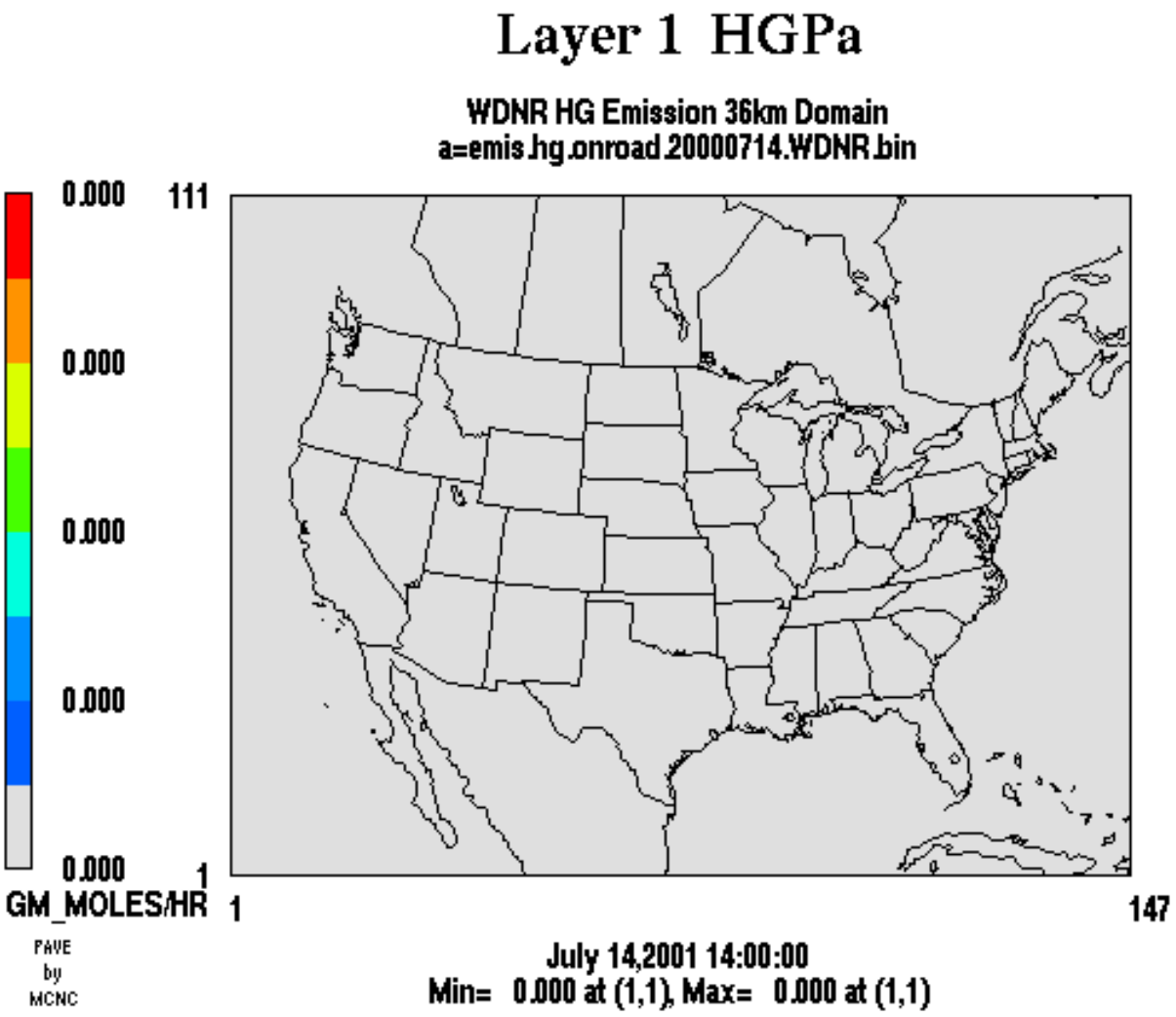


Figure 4-14. On-Road Motor Vehicle Spatial Allocation of HGP Emissions.

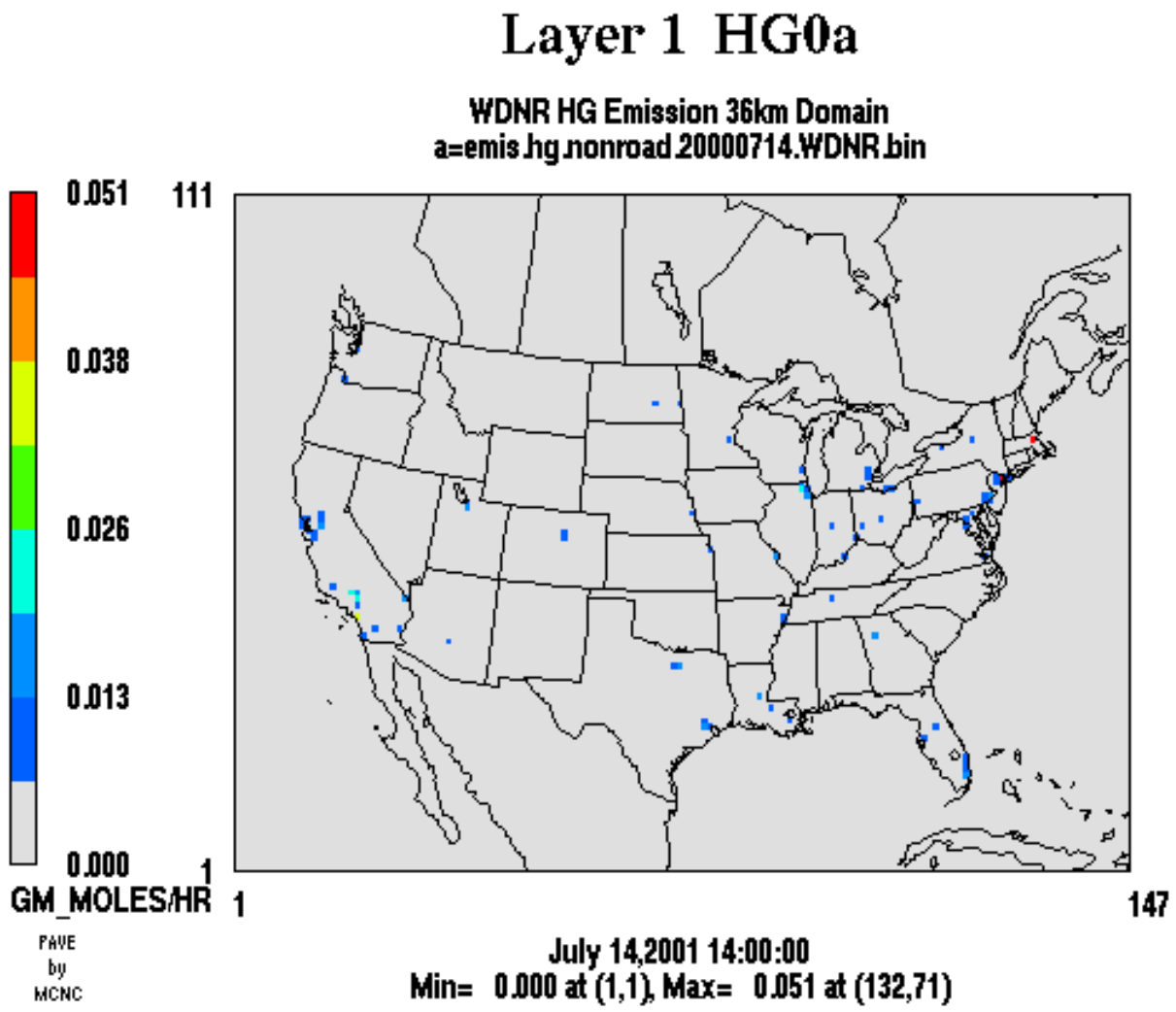


Figure 4-15. Non-Road Area Spatial Allocation of HG0 Emissions.

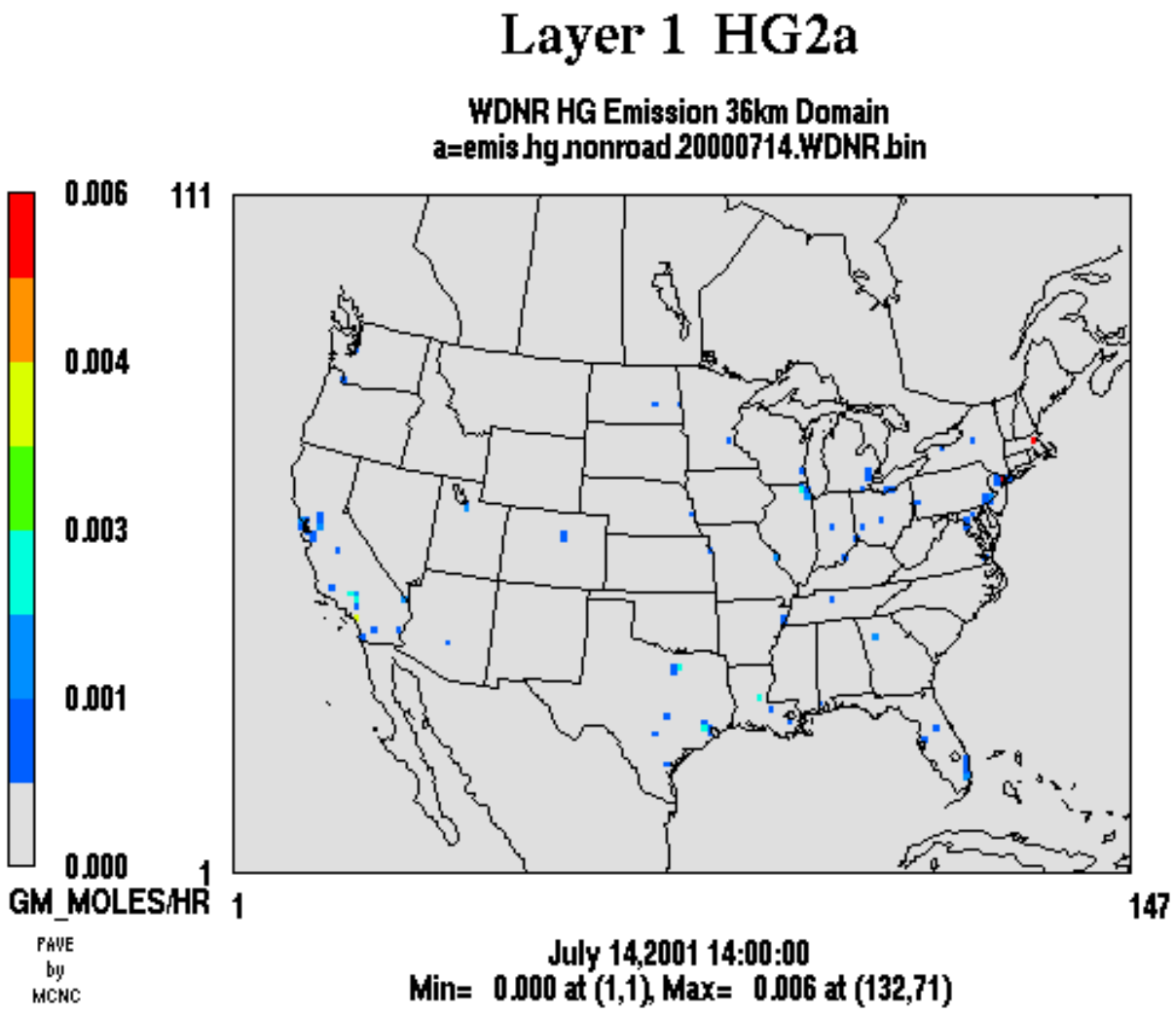


Figure 4-16. Non-Road Area Spatial Allocation of HG2 Emissions.

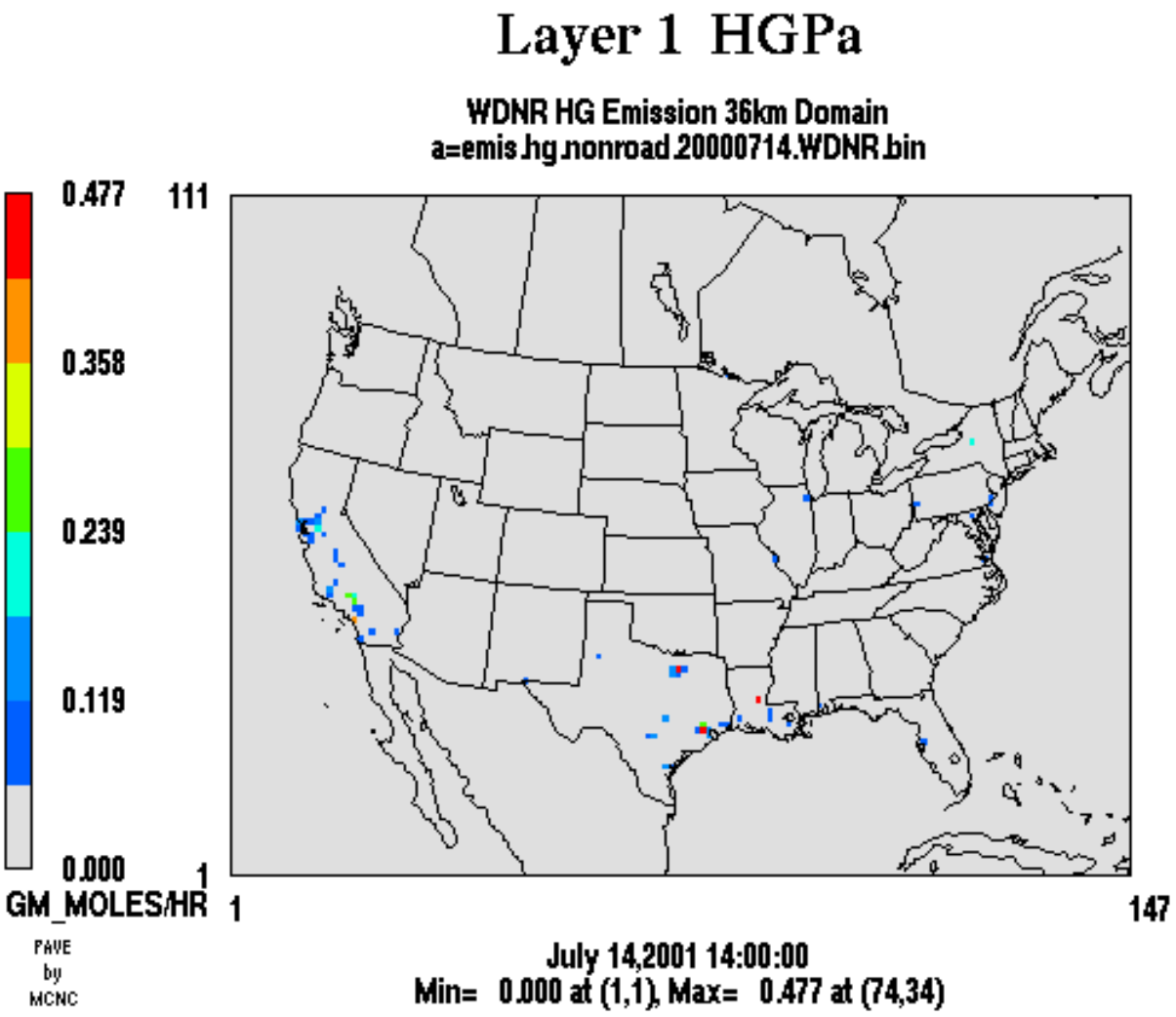


Figure 4-17. Non-Road Area Spatial Allocation of HGP Emissions.

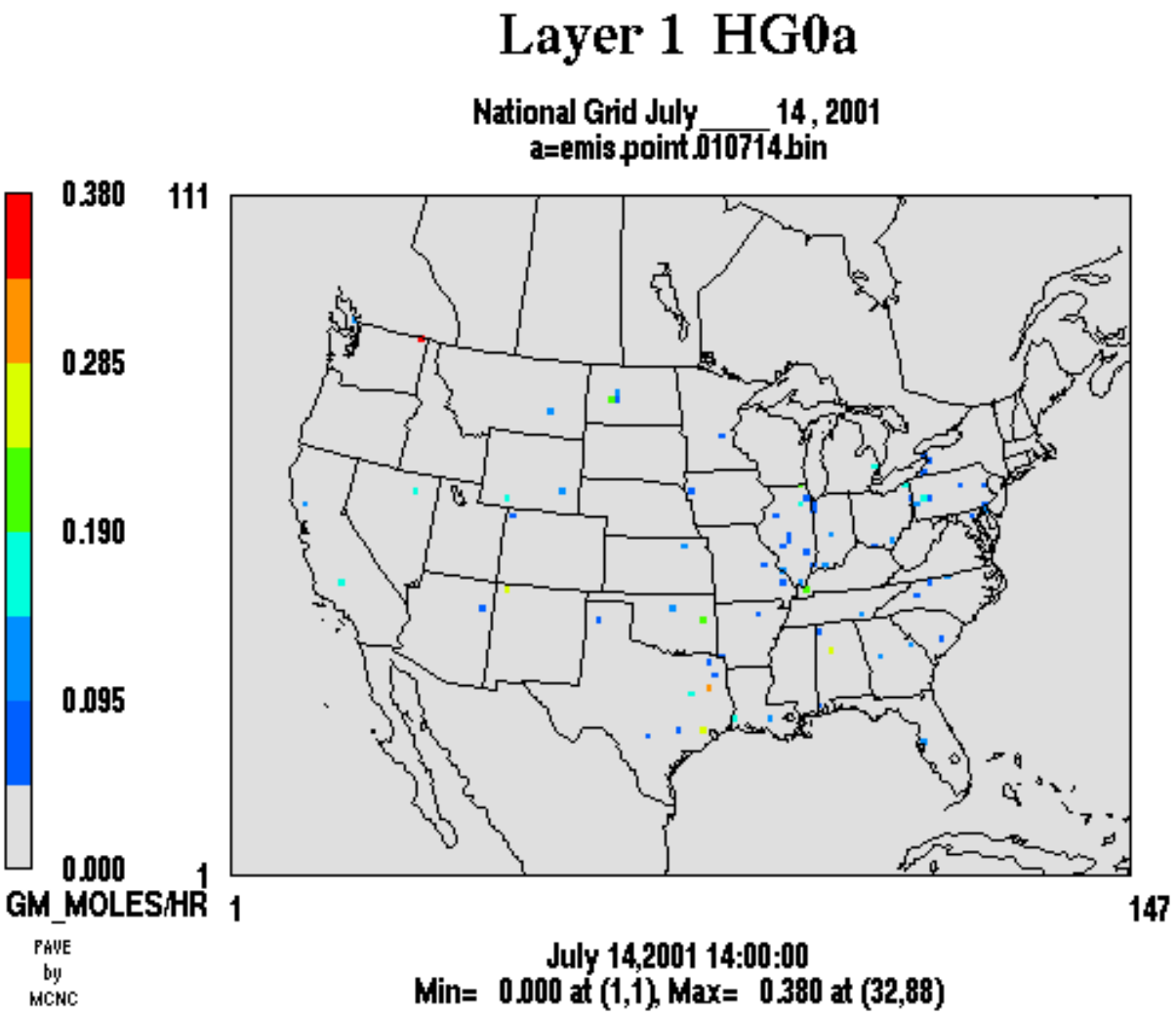


Figure 4-18. Elevated Point Source Spatial Allocation of HG0 Emissions.

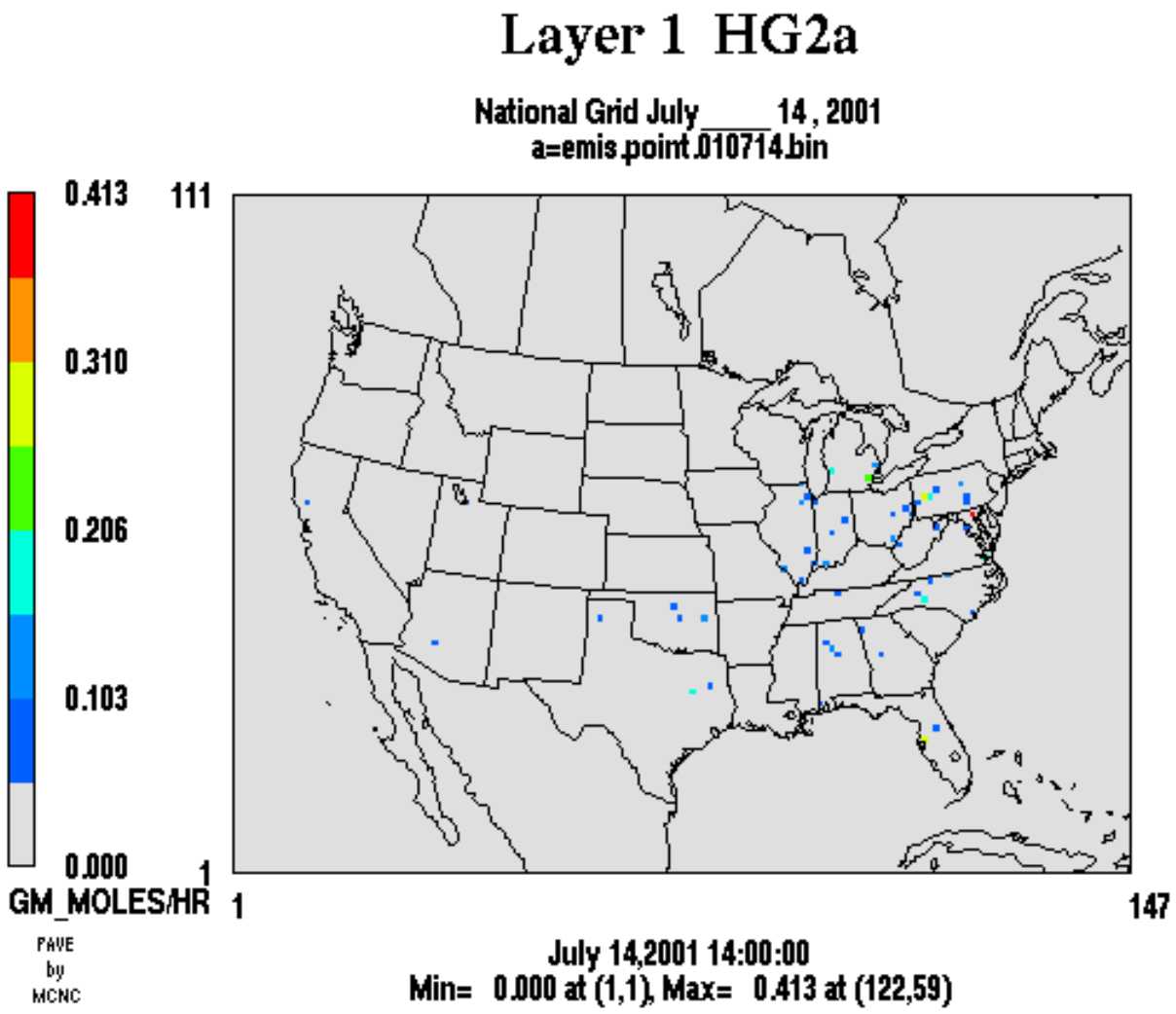


Figure 4-16. Elevated Point Source Spatial Allocation of HG2 Emissions.

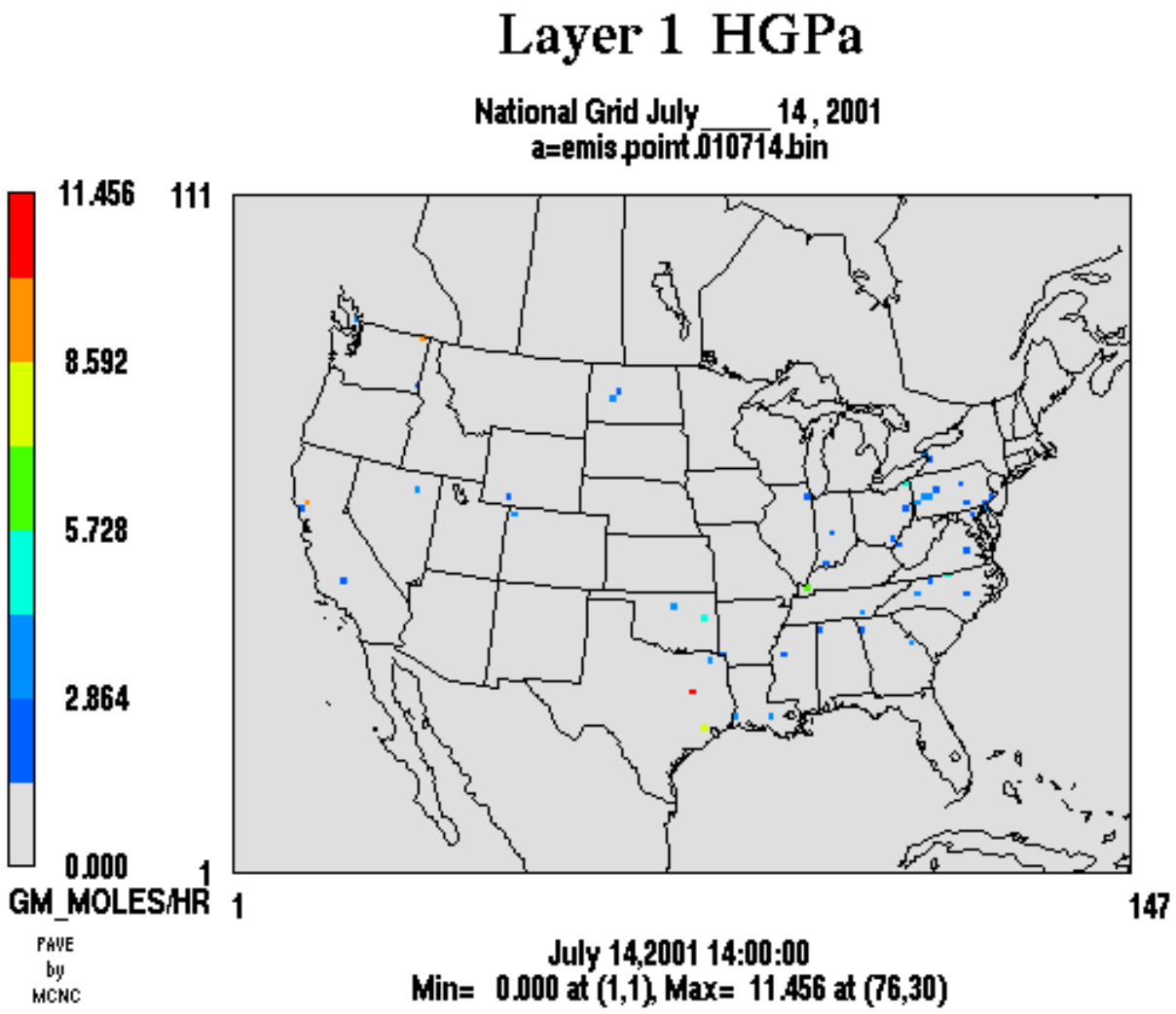


Figure 4-20. Elevated Point Source Spatial Allocation of HGP Emissions.

5.0 METEOROLOGICAL MODELING ASSESSMENT

A key component of the HgCAMx modeling system is the meteorological description of the three-dimensional, time dependent dynamic and thermodynamic variables of wind, temperature, mixing ratio, planetary boundary layer height, turbulent mixing rates, and so on. The meteorological inputs influence virtually all aspects for the modeling system, from the emissions rates for biogenic and motor vehicle emissions to the advection, diffusion and chemical transformation rates of gas phase and aerosol pollutants once they enter the atmosphere. Meteorology also has a profound effect on the removal of pollutant through wet and dry deposition processes. This section review the prognostic meteorological modeling methods used in support of the HgCAMx model development, initial application and performance testing. We also compare the meteorological model evaluation results presented by the WDNR and ENVIRON study teams with our own independent evaluations and the results derived from other contemporary annual modeling efforts.

5.1 Meteorological Modeling Approach

WDNR set up the MM5 prognostic model over the national RPO grid at 36 km scale (34 vertical layers) and carried out an annual simulation in order to support the HgCAMx model development and testing exercise. Data from the National Center for Environmental Prediction (NCEP) was used for initialization and data assimilation. Science options used in the annual simulation of the year 2002 included simple ice for moisture parameterization, the Kain-Fritsch cumulus parameterization, the Pleim-Xiu PBL scheme and soil model, and the Rapid Radiative Transfer Model (RRTM) radiation scheme. WDNR modelers invoked four dimensional data assimilation (FDDA), but only above the planetary boundary layer.

The WDNR MM5 application methodology is very similar to other recent studies, including the annual MM5 modeling performed by Alpine Geophysics for EPA Office of Air Quality Planning and Standards (McNally and Tesche, 2002, 2003). The WDNR approach is also comparable to three other recent modeling studies including: (a) the MM5 modeling work performed by Baron Advanced Meteorological Services (BAMS) in support of the VISTAS episodic and annual (2002) regional haze modeling (Olerud, 2003), (b) the 2002 MM5 application being performed for the Western Regional Air Partnership (WRAP) by UC Riverside (Wang, 2003), and (c) and the annual 2002 MM5 modeling performed by the Iowa Department of Natural Resources (IDNR) (Johnson, 2003). All of these modeling applications use identical horizontal and vertical modeling domains and data assimilation options. One difference, though, is that the WDNR used the Simple Ice moisture scheme while the other applications used higher order, mixed-phase moisture schemes. The choice of the simpler moisture scheme may be due to the fact that CAMx4 requires water vapor (in ppm), cloud water (in gm/m^3), and precipitable water (gm/m^3), but no estimates of solid phase precipitation.

5.2 Surface Temperature Evaluation

Temperature is important for mercury modeling since it influences the several source emissions rates, the rate of chemical transformation within the chemistry module, and the vertical temperature gradients which are important driving forces for winds. To independently assess the MM5 modeling portion of the HgCAMx exercise, and to gain insight into how the WNDR modeling compares with other studies, we analyzed the Layer 1 meteorological fields produced by the WNDR simulation for the entire year using the NCAR Co-Op (DS472.0) station archives. This evaluation used Alpine

Geophysics MAPS evaluation software package, a public domain code widely used to support performance evaluation of regional meteorological and air quality models.

Hourly surface temperature mean normalized bias and mean normalized error results for the WDNR simulation for two-month windows for the entire domain and for each state and RPO are presented in Tables 5-1 and 5-2, respectively. For the full year throughout the RPO region the mean normalized bias in hourly surface temperature estimation was –0.45 K while the mean gross error was 3.63 K. For comparison purposes, we list the recent EPA 2001 simulation bias and error estimates developed by McNally and Tesche (2003) in Tables 5-3 and 5-4. These results show that the mean bias and errors in the 2001 annual MM5 simulation were –0.51 K and 2.04 K, respectively. Results for the WRAP 2002 annual MM5 simulation (Wang, 2003) are presented in Tables 5-5 and 5-6. In the WRAP MM5 evaluation across the same domain, the hourly mean normalized bias and errors were –0.33 K and 1.62 K, respectively. Finally, based on over 60 MM5/RAMS model evaluations carried out by Alpine modelers since 1995 the mean bias and error in prognostic model simulations for regulatory modeling (at typical scales of 4 km to 12 km) 0.10 K and 2.0 K, respectively. While these historical comparisons provide a useful context for judging the WDNR, Alpine, and WRAP annual simulations, these were predominantly of episodic in nature, covered more limited areas and were developed in most cases using the best simulation of many different MM5 simulations. It would be impractical to iterate on an annual MM5 simulation to this degree, so it is hardly surprising that the WDNR annual simulation has somewhat poorer temperature bias and error scores compared with the national averages.

5.3 Surface Wind Evaluation

Proper specification of the three-dimensional wind fields is a crucial input for simulating the transport and removal of pollutants. We also exercised the AG MAPS evaluation software to independently evaluate the Layer 1 wind fields produced by MM5 for input to CAMx. In this analysis, we compared the CAMx-ready winds against available surface observations. In this type of comparison between model estimate and measurement, one must recognized that the MM5 modeled winds extracted from Layer 1 are valid, strictly speaking at a height of 18 meters compared with the nominal 10 meter observed anemometer heights. Even this modest difference in heights can lead to noticeable differences in the statistical quantities that reflect more on the challenges of obtaining direct comparisons between model and measurement at a common height rather than on inherent errors in the modeled and measured winds. Fortunately, all of the MM5 simulations we have examined in support of this peer review have identical vertical grid structures so model intercomparisons are useful nonetheless.

Wind speed index of agreement (I) results for the WDNR simulation for two-month windows for the entire domain and for each state and RPO are presented in Table 5-7. The annual mean index of agreement over the whole domain was 0.83 which are judged to be quite acceptable. Analogous index of agreement scores for the EPA 2001 simulation are presented in Table 5-8 with a mean value of 0.86. The index of agreement for the WRAP 2002 simulation (Table 5-9) yields a mean value of 0.89. The mean index of agreement in the same 60 studies described in the previous section showed was 0.72. On reason the mean I score for the historical studies is systematically lower than the 36 km annual runs is that the historical episodic simulations involved horizontal grid meshes typically of 4 km to 12 km and the index of agreement score systematically tends to higher values as the size of the horizontal grid mesh increases. Thus, while the index of agreement for the WNDR simulation is

somewhat worse than the other two comparison annual MM5 simulations, we do not believe this is cause for concern. Indeed, based on the surface wind and temperature evaluations reported above, we believe the MM5 simulations are quite adequate for prescribing input wind and temperature fields to the HgCAMx model at 36 km scale.

5.4 Precipitation Evaluation

An important class of input that influence the accuracy of HgCAMx's estimates of mercury transport, transformation, and deposition is the characterization of precipitation amount, timing, and location. From our previous experience in the RAMS/URM modeling for the Southern Appalachian Mountains Initiative (SAMI) project (Doty et al., 2002; Odman et al., 2002), we found that unless the dynamic and thermodynamic fields are reasonably well simulated, it is unlikely that the air quality model will reproduce the correct amounts of cloud and precipitation, and correspondingly, wet deposition. Unlike the three-dimensional wind and temperature estimates that are readily transferred from MM5 into HgCAMx (especially if the two models have the same vertical grid structures), proper 'mapping' of precipitation is more difficult. CAMx uses the three-dimensional rainwater variable for precipitable water content. To calculate the rainfall rate, the precipitable water content is then scaled by a water droplet fall velocity. The droplet velocity that used in the HgCAMx development and initial application effort has since been recognized by ENVIRON scientists to being too large, and a study is currently underway through LADCO to explore this issue in greater detail.

To explore just how the precipitation estimates developed by the WDNR MM5 simulation and used in the HgCAMx simulation relate to actual observations, we computed a metric referred to as the "accumulated precipitation bias". Accumulated precipitation is computed by summing the observed precipitation over the entire period (monthly in this case) at each station, and summing the model estimated precipitation at each station location over the same period. Once these monthly total precipitation values are calculated, the stations are then aggregated together statistically to produce standard bias and error metrics. By using the accumulated precipitation metric we were able to filter out the noise due to the exact timing of rainfall events and focus on rainfall trends. Our use of this comparison method does not diminish the need to examine the modeled vs. measured precipitation of individual events, but such an analysis was beyond the scope of this peer-review. Here, we have attempted to conduct a first-order review of the precipitation results, focusing primarily on time-integrated precipitation totals. Of course, this is only the first step in a thorough evaluation of the estimated precipitation fields; a full analysis of the WDNR MM5 simulation precipitation results is needed in order to clarify fully the adequacy of the precipitable water estimates supplied to HgCAMx.

Inter-comparisons of accumulated precipitation bias derived from the WDNR HgCAMx modeling and the Alpine's 2001 annual MM5 simulations for EPA is complicated by the fact that the WDNR precipitation was computed using the CAMx cloud/rain files and the MM5v3CAMxv4 methodology of computing rain based on the precipitable water content. The MM5 simulation we performed for EPA for 2001 used the MM5 output precipitation directly. Project resources precluded direct comparisons with MM5 fields, but since the HgCAMx precipitation is what is used in the mercury deposition, it is a more representative comparison.

Accumulated precipitation bias from the WDNR MM5 simulation of 2002 for the entire domain, and each state and RPO region are presented in Table 5-10. For the entire domain summed over the entire episode, the model is overestimating rainfall by 6.35 cm. Accumulated precipitation

error is presented in Table 5-11. The domain averaged episode total error is 7.48 cm. By contrast, the 2001 EPA annual accumulated precipitation bias and error are presented in Tables 5-12 and 5-13, respectively. The annual domain wide bias is 1.02 cm and 3.38 cm, respectively. To facilitate comparison of these two MM5 simulations, we developed time series plots of the mean monthly accumulated precipitation. The monthly precipitation time series for the WDNR annual simulation is presented in Figure 3-1 while the analogous plot for the EPA 2001 simulation appears in Figure 3-2. From this simple comparison, it is clear that the precipitation rates used in the HgCAMx modeling significantly overestimate the observed rainfall, a finding that was also discussed by Yarwood et al., (2003a). For the majority of the year, the modeled precipitable water supplied to HgCAMx was overestimated by approximately a factor of two, with the summer months being overestimated by a factor of 3.

As noted, this rainfall overestimation was recognized in the ENVIRON final report, and an appropriate HgCAMx model sensitivity exercised was performed to explore the potential implications of this bias on the modeled wet mercury deposition results. As reported by Yarwood et al., (2003a), the mercury wet deposition was reduced based on the ratio of the model estimated to observed precipitation. This scaling resulted in the mercury wet deposition values being much closer the MDN observations.

5.5 Synthesis of Meteorological Modeling Assessment

The modeling methodology used in the HgCAMx study was compared with other recent annual modeling studies and the approach is very similar. Surface temperatures were compared with observations and while the bias and error in this application are somewhat greater than the comparison WRAP and EPA simulations, the values are close and are judged acceptable. Wind speed index of agreement scores are also somewhat poorer than the other two studies, but overall they are quite consistent with other MM5 applications for air quality studies and are deemed acceptable.

The precipitation rates used in the HgCAMx model are much higher than observations (a factor of 2-3) and the performance is markedly inferior to the MM5 performance in comparison to the EPA annual modeling for 2001. Since mercury wet deposition is proportional to rainfall rate, this large error casts doubt on the present suitability of the HgCAMx modeling system and the 2002 episode as a credible planning tool for estimating wet mercury deposition. The model developer is well aware of this difficulty and is working to better understand the current limitations of both the MM5 meteorological model and the HgCAMx formulation. A suitable refined approach is expected. If it is critical to use the mercury wet deposition values estimated by HgCAMx, it is recommended that the rainfall rate scaled values be used. However, it is not clear whether the model's wet deposition bias is due just to precipitation problems or whether some other unknown compensating errors may be at play. According, the scaling suggestion is offered advisably.

Table 5-1. Temperature Bias (K) by State and Time Period for the WDNR 2002 Annual MM5 Simulation.

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
ALL	-0.43	-0.6	-0.49	-0.54	-0.23	-0.43	-0.45
AL	-0.12	-0.32	-0.34	-0.44	-0.32	0.02	-0.25
AZ	0.2	-0.18	-0.56	-1.43	-0.48	0.08	-0.4
AR	-0.23	-0.27	-0.03	-0.17	0.14	-0.09	-0.11
CA	0.36	-0.57	-1.04	-1.07	-0.43	0.05	-0.45
CO	-0.88	-1.8	-2.15	-1.92	-0.92	-1.1	-1.46
CT	-0.59	-0.37	-0.03	-0.01	0.17	-0.64	-0.24
DE	-0.18	0.1	-0.01	-0.38	-0.15	-0.48	-0.18
DC	-1.14	-0.57	-0.89	-1.53	-0.88	-1.2	-1.03
FL	-0.05	-0.54	-0.71	-0.58	-0.62	0.15	-0.39
GA	-0.03	-0.21	-0.25	-0.52	-0.22	0.15	-0.18
ID	-0.17	-1.37	-0.87	-0.49	-0.06	-0.46	-0.57
IL	-0.74	-0.4	0.13	-0.17	-0.04	-0.66	-0.31
IN	-1.04	-0.86	-0.32	-0.33	-0.14	-1.01	-0.62
IA	-0.74	-0.61	-0.25	-0.09	-0.05	-0.55	-0.38
KS	-0.57	-0.31	-0.38	-0.56	-0.09	-0.49	-0.4
KY	-0.69	-0.5	-0.21	-0.39	-0.04	-0.62	-0.41
LA	-0.24	-0.41	-0.59	-0.48	-0.33	0.26	-0.3
ME	-0.41	-0.76	-0.19	-0.02	0.01	-0.62	-0.33
MD	-0.04	0.07	-0.05	-0.44	0.03	-0.19	-0.1
MA	-0.32	-0.48	-0.26	-0.28	0.15	-0.33	-0.25
MI	-0.74	-0.73	-0.21	-0.14	-0.17	-0.86	-0.47
MN	-0.46	-0.65	-0.09	0.06	-0.31	-0.8	-0.38
MS	0.11	-0.09	-0.04	0.06	0.02	0.42	0.08
MO	-0.58	-0.49	-0.13	-0.31	0	-0.54	-0.34
MT	-1.01	-1.11	-1.17	-0.94	-0.73	-1.2	-1.03
NE	-0.51	-0.48	-0.48	-0.59	-0.24	-0.46	-0.46
NV	-0.57	-1.27	-1.38	-1.72	-0.64	-0.65	-1.04
NH	-0.23	-0.41	0.26	0.54	0.33	-0.33	0.03
NJ	-0.41	-0.29	-0.2	-0.36	0.15	-0.49	-0.27
NM	-0.45	-0.39	-1.66	-1.72	-0.53	-0.34	-0.85
NY	-0.77	-0.9	-0.41	-0.28	0	-0.68	-0.51
NC	-0.43	-0.36	-0.49	-0.71	-0.14	-0.24	-0.4
ND	-0.27	-0.67	-0.52	-0.24	-0.45	-0.57	-0.45
OH	-0.87	-0.71	-0.11	-0.3	0.01	-0.85	-0.47
OK	-0.47	-0.36	-0.33	-0.52	-0.04	-0.34	-0.34
OR	-0.84	-1.1	-0.82	-0.76	-0.18	-0.36	-0.68
PA	-0.96	-0.74	-0.23	-0.39	-0.05	-0.74	-0.52
RI	-0.35	-0.29	-0.25	-0.2	0.05	-0.37	-0.24
SC	-0.03	-0.25	-0.34	-0.78	-0.06	0.23	-0.2
SD	-0.7	-0.6	-0.4	-0.4	-0.33	-0.74	-0.53
TN	-0.5	-0.56	-0.23	-0.38	-0.09	-0.43	-0.37

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
TX	-0.05	-0.3	-0.7	-0.92	-0.35	-0.03	-0.39
UT	-0.03	-1.08	-0.83	-1.19	-0.11	-0.3	-0.59
VT	-0.91	-1.41	-0.51	0.04	-0.22	-1.13	-0.69
VA	-0.49	-0.27	-0.46	-0.78	-0.09	-0.31	-0.4
WA	-0.63	-0.76	-0.48	-0.24	0.02	-0.23	-0.39
WV	-1.12	-0.91	-0.1	-0.11	0.21	-0.8	-0.47
WI	-0.85	-0.68	0.07	-0.05	-0.25	-1	-0.46
WY	-1.09	-1.49	-1.7	-1.51	-0.97	-1.13	-1.32
CENRAP	-0.4	-0.45	-0.34	-0.39	-0.19	-0.39	-0.36
MANE_VU	-0.57	-0.61	-0.22	-0.21	0.04	-0.58	-0.36
MW	-0.82	-0.67	-0.08	-0.17	-0.14	-0.88	-0.46
VISTAS	-0.28	-0.39	-0.42	-0.55	-0.22	-0.08	-0.32
WRAP	-0.32	-0.87	-1.06	-1.06	-0.44	-0.41	-0.69

Table 5-2. Temperature Error (K) by State and Time Period for the WDNR 2002 Annual MM5 Simulation.

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
ALL	3.45	3.74	4.04	4.01	3.5	3.06	3.63
AL	3.63	3.85	3.66	3.38	2.78	3.16	3.41
AZ	5.06	5.65	6.21	5.23	4.89	4.47	5.25
AR	3.6	3.55	3.34	3.34	3.28	3.18	3.38
CA	3.79	3.98	4.59	4.95	4.5	3.36	4.2
CO	4.95	5.71	6.64	6.35	4.99	4.43	5.51
CT	2.91	3.25	3.5	3.49	2.97	2.52	3.11
DE	2.96	3.19	3.22	3.18	2.47	2.27	2.88
DC	3.73	3.78	3.95	3.81	3.11	2.77	3.53
FL	3.12	3.18	2.81	2.44	2.47	2.95	2.83
GA	3.92	3.97	3.82	3.59	2.82	3.27	3.57
ID	3.1	4.08	5.14	5.91	5.14	2.93	4.38
IL	3.06	3.34	3.48	3.56	3.66	2.79	3.31
IN	3.1	3.44	3.56	3.6	3.62	2.69	3.34
IA	3.46	3.9	4.11	3.66	3.67	3.27	3.68
KS	4.11	4.55	4.16	4.29	3.75	3.7	4.09
KY	3.23	3.55	3.36	3.46	3.08	2.44	3.19
LA	3.44	3.22	3.18	2.86	2.41	2.93	3.01
ME	2.83	2.82	3.51	3.66	3.13	2.59	3.09
MD	3.13	3.25	3.47	3.36	2.74	2.42	3.06
MA	2.69	2.94	3.16	3.33	2.82	2.48	2.9
MI	2.33	2.98	3.37	3.53	3.21	2.27	2.95
MN	3.03	3.26	3.9	3.4	3.04	2.86	3.25
MS	3.52	3.55	3.56	3.24	2.74	3.09	3.28
MO	3.5	3.83	3.51	3.62	3.57	3.26	3.55
MT	3.61	4.07	4.58	5.37	4.64	3.69	4.33
NE	4.34	4.41	4.79	4.72	3.99	4.02	4.38
NV	4.56	5.25	6.2	6.68	5.78	4.35	5.47
NH	3.55	3.89	4.36	4.55	3.89	3.27	3.92
NJ	3.15	3.37	3.55	3.6	2.97	2.51	3.19
NM	4.85	5.84	6.41	5.27	4.58	4.07	5.17
NY	2.73	3.06	3.21	3.4	2.88	2.28	2.93
NC	3.76	3.86	3.94	3.62	2.68	3.11	3.49
ND	3.12	3.57	4.3	4.14	3.47	2.88	3.58
OH	2.76	3.21	3.45	3.61	3.3	2.11	3.07
OK	4.24	4.11	3.81	3.95	3.5	3.63	3.87
OR	2.7	3.64	4.5	5.64	4.95	2.84	4.05
PA	2.97	3.48	3.68	3.82	3.17	2.32	3.24
RI	2.35	2.65	2.73	2.81	2.43	2.16	2.52
SC	3.67	3.73	3.67	3.62	2.55	2.95	3.37
SD	3.64	3.75	4.57	4.47	3.57	3.47	3.91
TN	3.49	3.78	3.59	3.4	2.87	2.77	3.32

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
TX	3.97	3.75	3.84	3.62	3.22	3.37	3.63
UT	3.87	4.63	5.96	6.04	4.56	3.43	4.75
VT	3.02	3.25	3.85	3.98	3.36	2.87	3.39
VA	3.7	3.88	3.94	3.72	2.98	2.83	3.51
WA	2.21	3.04	3.74	4.49	3.93	2.14	3.26
WV	3.51	3.94	3.79	3.62	3.28	2.49	3.44
WI	2.78	3.11	3.59	3.55	3.12	2.78	3.15
WY	4.22	4.35	5.53	5.98	4.53	3.83	4.74
CENRAP	3.66	3.76	3.89	3.67	3.34	3.29	3.6
MANE_VU	2.92	3.21	3.48	3.6	3.03	2.48	3.12
MW	2.71	3.16	3.48	3.56	3.31	2.5	3.12
VISTAS	3.55	3.68	3.57	3.31	2.75	2.97	3.31
WRAP	3.79	4.33	5.1	5.3	4.56	3.47	4.42

Table 5-3. Temperature Bias (K) by State and Time Period for the EPA 2001 Annual MM5 Simulation.

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
ALL	-0.66	-0.83	-0.68	-0.71	-0.09	-0.09	-0.51
AL	-0.62	-0.34	-0.25	-0.29	0.22	0.31	-0.16
AK	-1.29	-0.78	-1.16	-0.74	-0.67	-0.42	-0.84
AZ	-1.12	-1.64	-2.27	-2.92	-1.19	-0.13	-1.54
AR	-0.71	-0.28	0.32	-0.09	0.38	0.33	-0.01
CA	0.12	-1.01	-1.62	-1.56	-0.58	0.2	-0.74
CO	-1.5	-3.16	-2.57	-2.27	-0.96	-0.61	-1.84
CT	-1.24	-0.92	-0.14	-0.04	0.04	0.1	-0.37
DE	-0.32	-0.08	-0.36	-0.45	0.22	0.38	-0.1
DC	-1.82	-1.22	-1.16	-1.18	-1.26	-1.34	-1.33
FL	0.36	-0.42	-0.7	-0.63	-0.4	0.22	-0.26
GA	-0.39	-0.32	-0.1	-0.48	0.31	0.51	-0.08
ID	-0.88	-2.37	-1.59	-1.38	-0.27	-0.07	-1.09
IL	-0.87	-0.44	0.06	0.07	0.2	-0.11	-0.18
IN	-0.79	-0.45	-0.04	-0.02	0.09	-0.34	-0.26
IA	-0.51	-0.64	-0.24	-0.06	0.19	-0.25	-0.25
KS	-0.51	-0.3	-0.51	-0.65	0.1	-0.01	-0.31
KY	-1	-0.52	-0.13	-0.3	0.1	0.14	-0.28
LA	0.13	-0.19	-0.45	-0.44	0.12	0.33	-0.08
ME	-0.72	-1.15	0.02	-0.09	0.23	-0.25	-0.33
MD	-0.34	-0.28	-0.23	-0.25	0.14	0.23	-0.12
MA	-1.16	-0.86	-0.24	0.1	0.14	0.13	-0.31
MI	-0.93	-0.52	-0.24	-0.23	0.16	-0.3	-0.34
MN	-0.32	-0.57	0.12	0.41	0.25	-0.46	-0.09
MS	-0.23	-0.03	-0.06	-0.13	0.67	0.93	0.19
MO	-0.93	-0.41	-0.08	-0.19	0.13	-0.02	-0.25
MT	-1.02	-2.4	-2.1	-2.13	-1.04	-0.77	-1.58
NE	-0.55	-0.33	-0.61	-0.17	0.36	0.15	-0.19
NV	-1.76	-2.79	-3.15	-3.2	-0.76	-0.6	-2.04
NH	-0.7	-0.59	0.65	0.59	0.87	0.5	0.22
NJ	-1.15	-0.73	-0.55	-0.44	0.17	0.02	-0.45
NM	-1.12	-1.24	-2.24	-2.07	-0.61	-0.17	-1.24
NY	-1.1	-0.93	-0.46	-0.51	-0.12	-0.34	-0.58
NC	-0.59	-0.49	-0.68	-0.56	0.24	0.17	-0.32
ND	-0.11	-0.64	-0.06	-0.28	-0.11	-0.1	-0.22
OH	-1.11	-0.52	0.07	-0.11	0.04	-0.13	-0.29
OK	-0.52	-0.35	-0.38	-1.01	-0.16	-0.05	-0.41
OR	-0.91	-1.54	-1.57	-1.88	-0.49	-0.61	-1.17
PA	-1.32	-0.72	-0.2	-0.44	-0.11	-0.26	-0.51
RI	-1.11	-0.76	-0.57	0.09	0.05	-0.07	-0.4
SC	-0.34	-0.4	-0.6	-0.73	0.13	0.28	-0.28
SD	0.06	-0.44	-0.34	-0.27	0.04	-0.53	-0.25

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
TN	-0.98	-0.7	-0.23	-0.68	0.06	0.13	-0.4
TX	-0.45	-0.3	-0.92	-1.22	-0.21	0.06	-0.51
UT	-1.56	-2.51	-2.44	-2.73	-0.56	0.02	-1.63
VT	-1.63	-1.42	-0.22	-0.38	-0.09	-0.77	-0.75
VA	-0.82	-0.62	-0.5	-0.61	-0.14	-0.06	-0.46
WA	-0.73	-1.13	-0.9	-0.67	-0.12	-0.27	-0.64
WV	-1.04	-0.84	0.27	0.17	0.17	0.18	-0.18
WI	-1.14	-0.93	0.23	0.01	0.06	-0.57	-0.39
WY	-0.51	-2.66	-2.47	-2.69	-0.98	-0.68	-1.66
CENRAP	-0.46	-0.41	-0.33	-0.37	0.1	-0.08	-0.26
MANE_VU	-1.07	-0.83	-0.23	-0.23	0.08	-0.11	-0.4
MW	-0.99	-0.61	0	-0.08	0.12	-0.31	-0.31
VISTAS	-0.42	-0.46	-0.43	-0.5	0.05	0.25	-0.25
WRAP	-0.67	-1.62	-1.74	-1.75	-0.59	-0.24	-1.1

Table 5-4. Temperature Error (K) by State and Time Period for the EPA 2001 Annual MM5 Simulation.

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
ALL	2.06	2.00	2.07	2.10	2.01	2.00	2.04
AL	1.84	1.82	1.83	1.67	1.99	2.15	1.88
AK	1.77	1.54	1.42	1.21	1.19	1.63	1.46
AZ	2.44	2.81	3.40	4.10	3.32	2.57	3.11
AR	1.64	1.65	1.70	1.63	1.85	1.80	1.71
CA	2.43	2.32	2.89	2.80	2.67	2.25	2.56
CO	3.35	3.88	3.47	3.40	3.15	3.14	3.40
CT	2.01	1.76	1.59	1.57	1.77	1.67	1.73
DE	1.67	1.67	1.52	1.56	1.50	1.62	1.59
DC	2.11	1.55	1.43	1.46	1.47	1.50	1.59
FL	2.19	1.93	2.05	1.91	1.91	1.96	1.99
GA	1.92	1.82	1.80	1.71	1.96	2.09	1.88
ID	2.93	3.24	3.02	3.47	2.92	2.66	3.04
IL	1.61	1.53	1.62	1.67	1.53	1.43	1.57
IN	1.52	1.54	1.53	1.51	1.49	1.70	1.55
IA	1.73	1.61	1.63	1.62	1.70	1.69	1.66
KS	1.76	1.67	1.76	1.86	1.80	1.92	1.79
KY	1.68	1.56	1.67	1.60	1.59	1.81	1.65
LA	2.02	1.87	1.91	1.81	2.07	2.37	2.01
ME	2.00	2.05	1.73	1.68	1.69	1.55	1.78
MD	1.94	1.74	1.67	1.68	1.97	2.02	1.84
MA	2.12	1.86	1.79	1.66	1.86	1.73	1.84
MI	1.58	1.65	1.91	1.81	1.54	1.52	1.67
MN	1.90	1.86	1.78	1.81	1.63	1.73	1.78
MS	1.92	1.82	1.84	1.70	2.02	2.32	1.94
MO	1.70	1.55	1.54	1.58	1.72	1.62	1.62
MT	3.10	3.10	2.92	3.29	2.75	3.16	3.05
NE	1.94	1.72	1.90	1.92	1.86	2.20	1.92
NV	3.15	3.35	3.97	4.52	3.60	2.89	3.58
NH	2.76	2.78	2.59	2.61	2.69	2.33	2.63
NJ	2.15	1.66	1.68	1.76	1.94	1.82	1.84
NM	2.48	2.36	3.11	3.04	2.59	2.40	2.66
NY	2.01	1.78	1.71	1.84	1.82	1.78	1.82
NC	2.01	1.87	1.87	1.67	1.92	2.08	1.90
ND	1.88	1.80	1.63	1.88	1.81	2.15	1.86
OH	1.60	1.50	1.63	1.56	1.49	1.59	1.56
OK	1.68	1.63	1.67	1.90	1.84	1.85	1.76
OR	2.34	2.48	2.61	2.96	2.66	2.27	2.55
PA	1.93	1.66	1.66	1.69	1.64	1.67	1.71
RI	1.66	1.56	1.50	1.35	1.41	1.37	1.47
SC	1.80	1.77	1.76	1.61	1.76	1.93	1.77
SD	2.25	1.92	1.82	2.08	1.91	2.18	2.03

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
TN	1.82	1.69	1.73	1.62	1.77	1.97	1.77
TX	1.72	1.65	1.82	1.84	1.74	1.79	1.76
UT	3.07	3.19	3.44	3.99	2.97	2.51	3.19
VT	2.40	2.17	1.99	2.02	2.08	2.01	2.11
VA	2.13	2.01	1.95	1.85	2.12	2.24	2.05
WA	1.89	2.05	2.08	2.28	1.98	1.80	2.01
WV	1.99	1.89	1.86	1.63	1.88	2.20	1.91
WI	1.88	1.86	1.71	1.72	1.50	1.57	1.71
WY	3.14	3.23	3.22	3.79	2.86	3.14	3.23
CENRAP	1.79	1.71	1.76	1.79	1.76	1.83	1.77
MANE_VU	2.07	1.86	1.76	1.78	1.84	1.77	1.85
MW	1.66	1.65	1.73	1.70	1.52	1.55	1.64
VISTAS	2.00	1.86	1.89	1.74	1.93	2.08	1.92
WRAP	2.57	2.63	2.85	3.05	2.66	2.45	2.70

Table 5-5. Temperature Bias (K) by State and Time Period for the WRAP 2002 Annual MM5 Simulation.

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
ALL	-0.16	-0.35	-0.50	-0.60	-0.21	-0.14	-0.33
AL	0.11	-0.18	-0.26	-0.33	-0.20	0.15	-0.12
AK	0.44	-0.37	-1.06	-0.60	-0.23	-0.12	-0.32
AZ	0.33	-0.21	-0.78	-1.45	-0.49	0.23	-0.40
AR	0.13	0.02	0.11	-0.06	0.21	0.10	0.09
CA	0.63	-0.74	-1.95	-2.13	-0.95	0.50	-0.77
CO	-0.84	-1.77	-2.62	-2.36	-1.03	-1.05	-1.61
CT	-0.11	0.05	0.15	0.12	0.17	-0.30	0.01
DE	0.09	0.20	0.11	-0.36	-0.10	-0.18	-0.04
DC	-0.90	-0.46	-0.75	-1.68	-1.12	-1.09	-1.00
FL	0.15	-0.56	-0.71	-0.49	-0.52	0.31	-0.30
GA	0.12	-0.05	-0.14	-0.53	-0.17	0.08	-0.12
ID	-0.01	-1.51	-1.64	-0.72	-0.08	-0.27	-0.70
IL	-0.31	0.10	0.25	-0.20	-0.05	-0.19	-0.07
IN	-0.52	-0.42	-0.19	-0.28	-0.10	-0.59	-0.35
IA	-0.24	0.00	-0.01	0.14	0.14	0.08	0.02
KS	-0.09	0.07	-0.19	-0.50	-0.04	-0.06	-0.14
KY	-0.29	-0.14	-0.09	-0.38	-0.06	-0.30	-0.21
LA	-0.04	-0.33	-0.55	-0.42	-0.29	0.40	-0.20
ME	-0.23	-0.59	-0.19	0.01	0.02	-0.52	-0.25
MD	0.16	0.24	0.06	-0.52	-0.13	-0.15	-0.06
MA	-0.09	-0.10	-0.16	-0.21	0.11	-0.26	-0.12
MI	-0.48	-0.40	-0.21	-0.10	-0.10	-0.49	-0.30
MN	-0.18	-0.21	0.34	0.14	-0.09	-0.31	-0.05
MS	0.37	0.08	0.14	0.18	0.09	0.56	0.24
MO	-0.18	-0.02	-0.04	-0.27	0.00	-0.13	-0.11
MT	-1.09	-1.03	-1.79	-1.38	-0.77	-1.23	-1.22
NE	-0.02	-0.02	-0.39	-0.57	-0.07	0.00	-0.18
NV	-0.12	-1.05	-2.00	-1.74	-0.42	-0.25	-0.93
NH	0.03	0.06	0.61	0.62	0.42	-0.14	0.27
NJ	-0.28	-0.08	0.00	-0.33	-0.02	-0.41	-0.19
NM	0.11	0.08	-1.56	-1.62	-0.32	0.21	-0.52
NY	-0.61	-0.56	-0.45	-0.38	-0.10	-0.49	-0.43
NC	-0.19	-0.12	-0.37	-0.64	-0.16	-0.20	-0.28
ND	0.18	-0.32	-0.29	-0.18	-0.08	-0.01	-0.12
OH	-0.37	-0.15	0.10	-0.26	-0.01	-0.50	-0.20
OK	-0.16	-0.12	-0.08	-0.47	-0.01	-0.06	-0.15
OR	-0.90	-1.33	-1.53	-1.14	-0.36	-0.11	-0.90
PA	-0.54	-0.16	0.01	-0.28	-0.02	-0.33	-0.22
RI	0.04	0.04	-0.17	-0.10	0.13	-0.12	-0.03
SC	0.18	-0.01	-0.25	-0.69	0.02	0.29	-0.08
SD	-0.41	-0.16	-0.20	-0.35	-0.16	-0.26	-0.26

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
TN	-0.16	-0.37	-0.17	-0.35	-0.10	-0.17	-0.22
TX	0.24	0.06	-0.32	-0.79	-0.25	0.12	-0.16
UT	0.56	-0.45	-1.28	-0.99	0.32	0.45	-0.23
VT	-0.56	-0.95	-0.32	0.28	0.05	-0.71	-0.37
VA	-0.31	-0.10	-0.26	-0.80	-0.22	-0.34	-0.34
WA	-0.63	-0.84	-0.91	-0.52	-0.10	-0.10	-0.52
WV	-0.62	-0.23	0.22	-0.01	0.27	-0.27	-0.11
WI	-0.63	-0.23	0.44	0.07	-0.09	-0.51	-0.16
WY	-1.38	-1.69	-2.22	-2.08	-1.10	-1.29	-1.63
CENRAP	-0.04	-0.06	-0.07	-0.28	-0.07	-0.02	-0.09
MANE_VU	-0.31	-0.25	-0.10	-0.17	0.02	-0.37	-0.20
MW	-0.48	-0.24	0.09	-0.12	-0.08	-0.46	-0.21
VISTAS	-0.05	-0.21	-0.30	-0.49	-0.20	0.02	-0.20
WRAP	-0.13	-0.85	-1.57	-1.47	-0.55	-0.11	-0.78

Table 5-6. Temperature Error (K) by State and Time Period for the WRAP 2002 Annual MM5 Simulation.

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
ALL	1.60	1.57	1.83	1.74	1.47	1.49	1.62
AL	1.34	1.40	1.58	1.44	1.18	1.32	1.38
AK	1.62	1.34	1.46	1.03	1.15	1.27	1.31
AZ	1.77	1.87	2.34	2.63	2.15	1.74	2.08
AR	1.26	1.27	1.35	1.24	1.11	1.13	1.23
CA	2.38	2.30	2.85	2.99	2.53	2.30	2.56
CO	2.81	3.20	3.73	3.36	2.42	2.48	3.00
CT	1.35	1.20	1.39	1.33	1.15	1.35	1.29
DE	1.21	1.14	1.42	1.23	0.95	1.08	1.17
DC	1.35	1.02	1.25	1.80	1.28	1.33	1.34
FL	1.69	1.76	1.74	1.60	1.52	1.76	1.68
GA	1.40	1.42	1.55	1.48	1.16	1.28	1.38
ID	2.10	2.59	2.96	2.59	2.14	1.88	2.38
IL	1.16	1.10	1.35	1.21	1.13	1.01	1.16
IN	1.28	1.41	1.50	1.33	1.30	1.22	1.34
IA	1.33	1.17	1.40	1.19	1.10	1.12	1.22
KS	1.37	1.28	1.45	1.42	1.18	1.27	1.33
KY	1.19	1.22	1.34	1.28	1.07	0.95	1.17
LA	1.72	1.52	1.77	1.62	1.25	1.48	1.56
ME	1.23	1.29	1.38	1.31	1.20	1.25	1.28
MD	1.42	1.23	1.45	1.45	1.15	1.29	1.33
MA	1.45	1.39	1.45	1.49	1.32	1.45	1.43
MI	1.29	1.43	1.63	1.50	1.37	1.22	1.41
MN	1.33	1.27	1.55	1.27	1.16	1.18	1.29
MS	1.35	1.36	1.55	1.48	1.16	1.35	1.38
MO	1.18	1.18	1.24	1.24	1.13	1.09	1.18
MT	2.46	2.41	2.57	2.21	1.84	2.40	2.31
NE	1.66	1.35	1.75	1.64	1.28	1.60	1.55
NV	2.17	2.70	3.63	3.40	2.75	2.35	2.83
NH	2.06	2.24	2.30	2.14	1.93	2.12	2.13
NJ	1.43	1.25	1.45	1.46	1.19	1.38	1.36
NM	1.90	1.97	2.85	2.66	1.86	1.66	2.15
NY	1.57	1.46	1.58	1.54	1.35	1.43	1.49
NC	1.52	1.49	1.59	1.58	1.14	1.35	1.45
ND	1.48	1.22	1.74	1.40	1.24	1.33	1.40
OH	1.14	1.16	1.38	1.38	1.19	1.00	1.21
OK	1.51	1.30	1.33	1.37	1.22	1.28	1.33
OR	1.86	2.13	2.38	2.24	2.05	1.93	2.10
PA	1.39	1.25	1.43	1.42	1.19	1.16	1.31
RI	1.05	1.05	1.29	1.30	1.11	1.09	1.15
SC	1.36	1.34	1.35	1.39	0.97	1.16	1.26
SD	1.65	1.36	1.78	1.61	1.35	1.56	1.55

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
TN	1.26	1.34	1.41	1.35	1.02	1.06	1.24
TX	1.30	1.14	1.42	1.39	1.23	1.21	1.28
UT	2.16	2.30	3.10	2.96	2.19	1.81	2.42
VT	1.66	1.66	1.76	1.51	1.31	1.72	1.60
VA	1.64	1.47	1.65	1.68	1.31	1.49	1.54
WA	1.62	1.75	1.93	1.87	1.73	1.57	1.74
WV	1.70	1.50	1.56	1.36	1.25	1.18	1.43
WI	1.25	1.19	1.45	1.27	1.16	1.16	1.25
WY	3.02	2.68	3.15	3.05	2.25	2.73	2.81
CENRAP	1.37	1.25	1.47	1.35	1.18	1.23	1.31
MANE_VU	1.47	1.39	1.53	1.49	1.29	1.39	1.43
MW	1.23	1.27	1.49	1.36	1.24	1.14	1.29
VISTAS	1.51	1.50	1.59	1.53	1.24	1.40	1.46
WRAP	2.15	2.21	2.69	2.60	2.12	2.03	2.30

Table 5-7. Index of Agreement by State and Time Period for the WDNR 2002 Annual MM5 Simulation.

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
ALL	0.81	0.84	0.81	0.82	0.81	0.89	0.83
AL	0.75	0.79	0.55	0.52	0.47	0.67	0.63
AZ	0.62	0.61	0.63	0.61	0.65	0.53	0.61
AR	0.63	0.75	0.66	0.70	0.58	0.65	0.66
CA	0.74	0.69	0.72	0.64	0.67	0.85	0.72
CO	0.69	0.68	0.78	0.68	0.67	0.82	0.72
CT	0.45	0.50	0.50	0.51	0.40	0.50	0.48
DE	0.54	0.61	0.60	0.81	0.76	0.84	0.69
FL	0.64	0.70	0.71	0.55	0.60	0.70	0.65
GA	0.44	0.62	0.58	0.54	0.60	0.37	0.52
ID	0.56	0.72	0.54	0.66	0.72	0.79	0.67
IL	0.47	0.76	0.55	0.51	0.59	0.65	0.59
IN	0.46	0.68	0.44	0.52	0.60	0.45	0.52
IA	0.56	0.75	0.63	0.74	0.48	0.45	0.60
KS	0.64	0.67	0.56	0.73	0.56	0.59	0.62
KY	0.50	0.51	0.47	0.56	0.58	0.68	0.55
LA	0.75	0.54	0.60	0.68	0.69	0.56	0.64
ME	0.48	0.52	0.55	0.59	0.56	0.25	0.49
MD	0.21	0.41	0.51	0.54	0.63	0.58	0.48
MA	0.41	0.59	0.55	0.50	0.56	0.31	0.49
MI	0.52	0.45	0.60	0.66	0.63	0.61	0.58
MN	0.57	0.62	0.50	0.62	0.43	0.36	0.52
MS	0.75	0.68	0.54	0.51	0.44	0.61	0.59
MO	0.46	0.65	0.61	0.64	0.55	0.81	0.62
MT	0.56	0.62	0.54	0.54	0.70	0.71	0.61
NE	0.67	0.57	0.67	0.61	0.77	0.66	0.66
NV	0.70	0.66	0.66	0.69	0.75	0.84	0.72
NH	0.24	0.29	0.43	0.50	0.40	0.37	0.37
NJ	0.44	0.52	0.49	0.44	0.61	0.36	0.48
NM	0.65	0.48	0.78	0.74	0.64	0.53	0.64
NY	0.47	0.69	0.61	0.60	0.74	0.72	0.64
NC	0.56	0.50	0.61	0.59	0.68	0.53	0.58
ND	0.65	0.52	0.60	0.69	0.59	0.62	0.61
OH	0.50	0.47	0.59	0.57	0.56	0.62	0.55
OK	0.49	0.60	0.62	0.65	0.50	0.54	0.57
OR	0.63	0.76	0.59	0.67	0.80	0.78	0.70
PA	0.47	0.38	0.51	0.49	0.54	0.76	0.53
SC	0.36	0.53	0.59	0.32	0.65	0.62	0.51
SD	0.57	0.58	0.66	0.73	0.84	0.75	0.69
TN	0.59	0.52	0.45	0.52	0.67	0.64	0.56
TX	0.76	0.69	0.60	0.68	0.73	0.90	0.73
UT	0.62	0.68	0.63	0.66	0.70	0.84	0.69

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
VT	0.38	0.38	0.49	0.46	0.47	0.57	0.46
VA	0.54	0.51	0.50	0.59	0.63	0.64	0.57
WA	0.60	0.74	0.61	0.77	0.73	0.87	0.72
WV	0.47	0.43	0.42	0.57	0.58	0.56	0.50
WI	0.56	0.49	0.46	0.54	0.51	0.20	0.46
WY	0.58	0.81	0.48	0.58	0.70	0.60	0.63
CENRAP	0.87	0.87	0.75	0.81	0.78	0.86	0.82
MANE_VU	0.48	0.62	0.63	0.64	0.70	0.73	0.63
MW	0.56	0.65	0.62	0.67	0.66	0.70	0.64
VISTAS	0.75	0.74	0.71	0.62	0.70	0.73	0.71
WRAP	0.78	0.78	0.77	0.80	0.85	0.87	0.81

Table 5-8. Index of Agreement by State and Time Period for the EPA 2001 Annual MM5 Simulation.

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
ALL	0.88	0.86	0.85	0.89	0.85	0.85	0.86
AL	0.72	0.73	0.75	0.71	0.61	0.75	0.71
AK	0.64	0.36	0.44	0.52	0.49	0.59	0.51
AZ	0.71	0.72	0.64	0.71	0.82	0.68	0.71
AR	0.73	0.64	0.65	0.76	0.65	0.73	0.69
CA	0.87	0.77	0.80	0.81	0.82	0.74	0.80
CO	0.70	0.77	0.70	0.80	0.71	0.75	0.74
CT	0.47	0.58	0.50	0.46	0.60	0.52	0.52
DE	0.61	0.78	0.81	0.92	0.86	0.81	0.80
FL	0.77	0.60	0.57	0.71	0.68	0.66	0.67
GA	0.65	0.64	0.55	0.54	0.57	0.69	0.61
ID	0.69	0.74	0.76	0.80	0.59	0.60	0.70
IL	0.69	0.63	0.65	0.69	0.53	0.54	0.62
IN	0.69	0.62	0.52	0.69	0.59	0.62	0.62
IA	0.73	0.64	0.70	0.62	0.44	0.41	0.59
KS	0.74	0.72	0.72	0.72	0.72	0.66	0.71
KY	0.70	0.47	0.56	0.47	0.44	0.55	0.53
LA	0.85	0.65	0.77	0.74	0.63	0.62	0.71
ME	0.45	0.57	0.50	0.50	0.57	0.50	0.51
MD	0.68	0.56	0.55	0.47	0.61	0.55	0.57
MA	0.50	0.63	0.58	0.34	0.54	0.45	0.51
MI	0.64	0.64	0.58	0.72	0.63	0.69	0.65
MN	0.62	0.72	0.74	0.65	0.71	0.56	0.67
MS	0.75	0.67	0.65	0.68	0.48	0.55	0.63
MO	0.72	0.68	0.64	0.74	0.67	0.64	0.68
MT	0.76	0.73	0.72	0.83	0.80	0.64	0.75
NE	0.81	0.80	0.73	0.85	0.71	0.73	0.77
NV	0.60	0.74	0.77	0.68	0.73	0.73	0.71
NH	0.23	0.37	0.28	0.20	0.34	0.12	0.26
NJ	0.40	0.54	0.55	0.53	0.57	0.48	0.51
NM	0.83	0.77	0.70	0.73	0.73	0.83	0.77
NY	0.64	0.74	0.65	0.56	0.71	0.55	0.64
NC	0.61	0.61	0.60	0.58	0.57	0.65	0.60
ND	0.52	0.64	0.68	0.85	0.72	0.59	0.67
OH	0.60	0.54	0.49	0.57	0.60	0.71	0.59
OK	0.53	0.69	0.57	0.55	0.70	0.66	0.62
OR	0.73	0.72	0.74	0.84	0.71	0.79	0.76
PA	0.51	0.72	0.56	0.60	0.71	0.68	0.63
RI	0.47	0.66	0.69	0.50	0.68	0.61	0.60
SC	0.61	0.73	0.53	0.58	0.48	0.55	0.58
SD	0.71	0.84	0.67	0.83	0.65	0.69	0.73

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
TN	0.67	0.59	0.59	0.56	0.45	0.57	0.57
TX	0.86	0.70	0.72	0.75	0.71	0.79	0.76
UT	0.67	0.73	0.77	0.65	0.71	0.61	0.69
VT	0.62	0.50	0.53	0.52	0.47	0.39	0.51
VA	0.76	0.58	0.62	0.59	0.64	0.67	0.64
WA	0.68	0.66	0.78	0.79	0.69	0.76	0.73
WV	0.56	0.55	0.53	0.50	0.59	0.53	0.54
WI	0.56	0.54	0.61	0.56	0.66	0.54	0.58
WY	0.77	0.63	0.84	0.79	0.62	0.66	0.72
CENRAP	0.93	0.82	0.87	0.88	0.80	0.85	0.86
MANE_VU	0.49	0.68	0.57	0.62	0.72	0.50	0.60
MW	0.75	0.73	0.64	0.80	0.69	0.68	0.71
VISTAS	0.84	0.81	0.77	0.74	0.76	0.78	0.78
WRAP	0.87	0.83	0.88	0.88	0.82	0.83	0.85

Table 5-9. Index of Agreement by State and Time Period for the WRAP 2002 Annual MM5 Simulation.

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
ALL	0.90	0.91	0.89	0.88	0.86	0.91	0.89
AL	0.84	0.86	0.69	0.63	0.63	0.79	0.74
AK	0.50	0.70	0.46	0.66	0.26	0.41	0.50
AZ	0.76	0.84	0.76	0.70	0.79	0.78	0.77
AR	0.83	0.81	0.78	0.75	0.69	0.76	0.77
CA	0.84	0.82	0.78	0.69	0.74	0.72	0.77
CO	0.81	0.80	0.86	0.79	0.77	0.74	0.79
CT	0.53	0.58	0.46	0.56	0.61	0.55	0.55
DE	0.67	0.89	0.90	0.93	0.91	0.76	0.84
FL	0.72	0.77	0.73	0.66	0.75	0.61	0.71
GA	0.66	0.74	0.65	0.60	0.69	0.71	0.68
ID	0.71	0.75	0.69	0.69	0.74	0.73	0.72
IL	0.64	0.86	0.59	0.67	0.65	0.71	0.69
IN	0.60	0.80	0.51	0.63	0.67	0.64	0.64
IA	0.68	0.86	0.76	0.78	0.69	0.59	0.73
KS	0.82	0.84	0.70	0.84	0.69	0.82	0.79
KY	0.68	0.69	0.61	0.63	0.65	0.73	0.67
LA	0.80	0.72	0.64	0.70	0.79	0.67	0.72
ME	0.53	0.65	0.58	0.72	0.59	0.54	0.60
MD	0.34	0.63	0.51	0.73	0.75	0.62	0.60
MA	0.54	0.69	0.61	0.55	0.67	0.48	0.59
MI	0.72	0.69	0.71	0.72	0.72	0.83	0.73
MN	0.71	0.75	0.76	0.72	0.63	0.64	0.70
MS	0.80	0.74	0.62	0.61	0.61	0.69	0.68
MO	0.67	0.76	0.75	0.70	0.69	0.68	0.71
MT	0.79	0.78	0.82	0.67	0.76	0.78	0.77
NE	0.83	0.83	0.81	0.75	0.85	0.75	0.80
NV	0.82	0.81	0.77	0.82	0.84	0.76	0.80
NH	0.26	0.34	0.40	0.56	0.45	0.18	0.37
NJ	0.57	0.61	0.50	0.48	0.60	0.49	0.54
NM	0.88	0.68	0.83	0.80	0.79	0.88	0.81
NY	0.70	0.82	0.66	0.70	0.82	0.67	0.73
NC	0.67	0.72	0.65	0.57	0.69	0.56	0.64
ND	0.77	0.71	0.84	0.86	0.77	0.68	0.77
OH	0.62	0.76	0.61	0.62	0.62	0.76	0.67
OK	0.76	0.78	0.72	0.74	0.64	0.65	0.71
OR	0.75	0.85	0.76	0.77	0.85	0.72	0.78
PA	0.70	0.71	0.53	0.63	0.68	0.74	0.67
RI	0.53	0.57	0.48	0.45	0.60	0.56	0.53
SC	0.57	0.73	0.70	0.42	0.68	0.51	0.60
SD	0.78	0.77	0.90	0.87	0.89	0.71	0.82
TN	0.69	0.70	0.53	0.68	0.75	0.76	0.68

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
TX	0.90	0.79	0.70	0.78	0.74	0.76	0.78
UT	0.77	0.80	0.76	0.77	0.83	0.75	0.78
VT	0.46	0.48	0.40	0.57	0.44	0.55	0.48
VA	0.64	0.71	0.55	0.63	0.67	0.66	0.64
WA	0.77	0.84	0.75	0.83	0.77	0.72	0.78
WV	0.49	0.64	0.51	0.58	0.57	0.62	0.57
WI	0.64	0.77	0.63	0.64	0.59	0.59	0.64
WY	0.75	0.86	0.74	0.76	0.81	0.70	0.77
CENRAP	0.94	0.93	0.85	0.86	0.86	0.87	0.88
MANE_VU	0.63	0.78	0.68	0.75	0.76	0.68	0.71
MW	0.75	0.85	0.72	0.74	0.74	0.82	0.77
VISTAS	0.85	0.86	0.76	0.68	0.76	0.89	0.80
WRAP	0.88	0.88	0.88	0.87	0.89	0.85	0.87

Table 5-10. Accumulated Precipitation Bias (cm) by State and Time Period for the WDNR 2002 Annual MM5 Simulation.

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
ALL	5.83	7.02	7.59	11.20	2.34	4.14	6.35
AL	1.96	4.97	8.84	18.04	4.13	-3.42	5.75
AZ	0.57	1.40	0.83	13.73	3.83	1.61	3.66
AR	1.75	2.99	6.34	14.98	1.71	3.18	5.16
CA	0.78	0.87	0.24	2.50	0.43	1.90	1.12
CO	7.11	6.57	7.01	7.95	2.83	4.24	5.95
CT	7.86	6.37	4.19	5.35	-2.63	14.85	6.00
DE	5.36	4.22	5.55	11.19	5.71	8.16	6.70
FL	0.27	4.53	10.03	20.39	12.39	-2.89	7.45
GA	1.54	5.28	12.52	19.84	1.99	-1.52	6.61
ID	18.43	11.70	5.25	4.88	2.43	9.23	8.65
IL	5.67	7.09	7.85	10.23	2.21	3.92	6.16
IN	5.76	8.74	12.09	13.07	3.83	6.21	8.28
IA	4.98	6.62	8.46	11.57	3.33	1.39	6.06
KS	6.71	3.83	6.86	10.64	3.13	3.24	5.74
KY	5.19	9.26	15.42	16.86	5.44	6.91	9.85
LA	0.96	0.83	10.18	20.28	-0.42	-4.33	4.58
ME	17.75	13.25	5.53	3.97	2.18	17.04	9.95
MD	5.43	8.14	9.56	10.70	2.91	9.94	7.78
MA	11.58	7.75	2.04	5.23	0.96	13.17	6.79
MI	10.65	11.85	6.95	6.78	3.13	4.10	7.24
MN	6.49	12.36	6.18	8.06	2.47	3.02	6.43
MS	-0.67	3.73	9.89	20.28	-0.54	-1.79	5.15
MO	5.96	6.01	3.35	9.99	2.89	6.07	5.71
MT	13.06	15.00	10.54	6.65	3.40	6.02	9.11
NE	4.38	7.12	8.23	8.21	3.35	1.49	5.46
NV	3.79	3.45	3.77	3.70	2.58	4.21	3.58
NH	15.16	19.12	5.30	3.70	0.48	22.46	11.04
NJ	4.45	4.03	8.49	11.56	2.08	8.09	6.45
NM	1.89	1.32	10.36	18.31	3.23	2.53	6.27
NY	13.37	12.47	5.52	6.05	0.00	18.76	9.36
NC	5.14	7.60	11.52	13.91	-1.05	5.08	7.03
ND	5.47	9.03	9.02	9.21	3.01	5.08	6.80
OH	5.79	8.85	9.45	16.84	3.45	6.87	8.54
OK	3.88	4.38	7.55	11.65	1.52	4.85	5.64
OR	4.74	4.28	1.19	0.86	-0.31	0.35	1.85
PA	9.12	9.40	9.53	8.72	2.59	12.84	8.70
RI	8.14	2.20	3.64	6.81	-0.46	9.29	4.94
SC	3.41	6.07	12.11	16.63	-2.56	1.81	6.25
SD	4.51	9.68	6.32	8.17	3.52	1.67	5.64
TN	2.78	4.54	9.66	14.90	2.31	1.37	5.93
TX	0.57	3.24	9.32	16.63	0.48	-0.36	4.98

UT	6.10	7.73	2.64	6.99	2.47	4.60	5.09
VT	15.62	20.01	6.14	5.37	0.68	25.07	12.15
VA	6.82	6.62	10.69	15.24	1.57	8.04	8.16
WA	14.94	8.92	2.14	0.39	-0.49	4.84	5.12
WV	9.93	10.49	13.96	9.08	6.82	11.91	10.36
WI	7.75	15.74	5.71	7.40	1.69	1.89	6.70
WY	8.22	12.69	7.28	9.17	5.80	6.16	8.22
CENRAP	3.50	5.02	7.61	13.01	1.88	1.94	5.49
MANE_VU	11.90	11.54	6.96	7.06	1.51	15.92	9.15
MW	6.83	10.18	8.64	11.20	2.85	4.76	7.41
VISTAS	3.36	6.11	11.24	16.96	3.22	1.96	7.14
WRAP	6.53	6.63	5.19	7.29	2.29	3.61	5.26

Table 5-11. Accumulated Precipitation Error (cm) by State and Time Period for the WDNR 2002 Annual MM5 Simulation.

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
ALL	6.62	7.74	8.06	11.58	4.58	6.29	7.48
AL	3.59	6.50	9.29	18.07	6.70	4.51	8.11
AZ	1.32	1.68	0.94	13.79	4.98	2.02	4.12
AR	3.08	4.86	7.33	15.15	3.75	4.76	6.49
CA	3.79	3.20	1.04	2.67	0.88	10.00	3.60
CO	7.11	6.58	7.03	8.27	3.64	4.25	6.15
CT	7.86	6.42	4.33	6.80	2.90	14.85	7.19
DE	5.36	4.22	5.55	11.19	6.29	8.16	6.80
FL	2.78	7.42	11.36	20.95	13.42	5.37	10.22
GA	2.45	6.38	12.54	19.84	6.53	3.23	8.49
ID	18.43	11.70	5.26	5.17	2.51	9.36	8.74
IL	5.72	7.24	8.29	10.67	3.53	4.15	6.60
IN	5.76	8.81	12.09	13.32	4.14	6.38	8.42
IA	4.98	7.11	8.52	12.17	4.11	1.48	6.39
KS	6.73	4.17	8.46	10.64	4.19	3.36	6.26
KY	5.19	9.26	15.43	16.86	5.81	7.10	9.94
LA	2.25	3.93	10.49	21.32	13.13	4.95	9.35
ME	17.75	13.25	5.63	5.38	4.51	17.04	10.59
MD	5.43	8.14	9.56	10.70	3.97	10.22	8.00
MA	11.58	7.82	2.83	6.01	2.48	13.17	7.32
MI	10.65	11.85	7.25	7.38	3.80	4.17	7.52
MN	6.49	12.36	6.54	9.26	2.98	3.04	6.78
MS	2.85	4.92	10.22	21.17	6.70	3.73	8.27
MO	6.02	6.32	5.78	10.52	4.28	6.19	6.52
MT	13.06	15.00	10.63	6.76	3.50	6.03	9.16
NE	4.38	7.49	8.50	9.18	3.86	1.53	5.82
NV	3.80	3.57	3.92	3.80	2.63	4.43	3.69
NH	15.16	19.12	7.05	4.24	3.75	22.46	11.96
NJ	4.45	4.12	8.49	11.56	3.35	8.47	6.74
NM	2.40	1.59	10.36	18.31	3.85	2.92	6.57
NY	13.37	12.51	5.68	6.80	2.98	18.76	10.02
NC	5.15	7.65	11.60	14.60	4.22	6.27	8.25
ND	5.47	9.03	9.22	9.41	3.24	5.08	6.91
OH	5.79	8.95	9.56	16.84	3.88	6.90	8.65
OK	4.18	5.13	8.11	11.75	5.04	5.22	6.57
OR	7.73	5.87	2.14	1.09	1.05	6.50	4.06
PA	9.12	9.51	9.66	8.79	4.47	12.97	9.09
RI	8.14	2.20	3.64	6.81	3.00	9.29	5.51
SC	3.44	6.14	12.26	18.01	5.62	3.86	8.22
SD	4.51	9.68	6.41	8.23	3.81	1.69	5.72
TN	4.17	7.79	10.16	15.43	5.35	2.72	7.60
TX	1.87	4.08	9.94	17.03	8.55	2.73	7.37

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
UT	6.23	7.73	2.89	7.03	3.58	4.66	5.35
VT	15.62	20.01	6.41	5.37	4.49	25.07	12.83
VA	6.89	6.65	10.91	15.95	3.92	8.83	8.86
WA	16.64	11.07	3.76	1.29	1.42	9.33	7.25
WV	9.93	10.49	14.02	9.52	7.12	11.91	10.50
WI	7.75	15.74	5.81	7.62	3.11	1.94	6.99
WY	8.22	12.69	7.28	9.17	5.95	6.16	8.24
CENRAP	4.10	5.81	8.40	13.51	5.83	3.61	6.88
MANE_VU	11.90	11.60	7.24	7.52	3.84	15.99	9.68
MW	6.84	10.25	8.82	11.48	3.69	4.87	7.66
VISTAS	4.52	7.18	11.57	17.47	6.70	5.49	8.82
WRAP	7.81	7.55	5.66	7.50	2.97	6.65	6.36

Table 5-12. Accumulated Precipitation Bias (cm) by State and Time Period for the EPA 2001 Annual MM5 Simulation.

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
ALL	-0.31	0.23	1.58	4.48	0.20	-0.06	1.02
AL	-2.19	-3.99	0.61	5.52	2.17	-2.00	0.02
AK	-2.09	-0.19	-5.04	-2.30	-1.84	1.70	-1.63
AZ	2.01	2.08	3.00	9.25	2.86	0.79	3.33
AR	-3.50	-1.19	1.01	9.10	-1.69	-2.29	0.24
CA	-3.77	-1.17	0.04	1.91	-0.13	-3.11	-1.04
CO	0.54	0.34	2.45	0.26	0.08	0.31	0.66
CT	0.16	0.18	-1.36	-0.16	-1.23	1.13	-0.21
DE	0.31	-1.57	-3.37	7.38	-0.44	0.02	0.39
FL	0.32	1.39	7.53	18.32	10.31	1.56	6.57
GA	-1.24	0.00	0.24	9.13	2.39	-0.41	1.69
ID	0.54	0.63	1.36	2.87	0.79	3.54	1.62
IL	0.35	0.31	0.85	5.53	-0.77	0.71	1.16
IN	-0.33	1.60	3.60	2.73	-0.96	1.42	1.34
IA	0.60	0.61	-0.31	4.01	-1.60	0.49	0.63
KS	-0.18	1.08	1.13	6.27	0.94	0.26	1.58
KY	-0.38	1.28	8.70	13.60	-0.51	0.34	3.84
LA	-1.53	-2.93	0.67	6.51	0.36	-1.40	0.28
ME	0.43	0.72	0.36	1.51	-2.21	0.60	0.23
MD	1.52	0.25	-1.57	-0.70	-1.03	0.24	-0.21
MA	0.69	1.80	-1.18	-0.63	-1.01	0.71	0.06
MI	0.59	-0.04	-0.16	1.64	-1.06	0.95	0.32
MN	1.11	-0.27	0.35	2.34	-0.43	0.44	0.59
MS	-4.16	-2.70	3.35	9.38	1.47	-2.07	0.88
MO	-1.08	0.86	-1.24	7.13	-1.16	-0.34	0.69
MT	1.00	1.56	2.68	2.86	0.77	1.63	1.75
NE	0.75	-0.28	3.64	2.16	-1.42	0.54	0.90
NV	1.01	1.66	1.42	5.01	1.53	1.94	2.10
NH	-0.88	1.58	-1.38	-0.74	-3.20	0.85	-0.63
NJ	0.67	-1.76	-2.02	2.65	0.50	0.63	0.11
NM	0.96	1.54	4.98	5.97	0.86	-0.03	2.38
NY	0.56	0.93	-1.06	0.25	-1.60	1.09	0.03
NC	-0.03	0.77	8.10	7.67	-0.55	-0.05	2.65
ND	0.56	0.42	0.38	1.39	0.47	0.37	0.60
OH	-0.02	0.53	1.98	4.41	-0.54	2.06	1.40
OK	-0.19	0.43	-0.20	4.85	-0.44	-0.27	0.70
OR	-0.65	-0.49	-0.35	0.61	-0.16	-1.22	-0.38
PA	0.29	-0.45	1.07	1.95	-1.21	0.38	0.34
RI	-0.87	-0.45	-3.41	-2.56	-1.71	2.13	-1.14
SC	-0.80	1.73	6.42	14.31	0.84	0.13	3.77
SD	1.28	0.57	0.37	2.05	0.00	0.83	0.85
TN	-3.42	-0.12	3.65	12.20	-0.02	-3.00	1.55
TX	1.08	1.32	2.79	5.86	2.79	-0.81	2.17
UT	0.88	0.53	2.81	4.41	1.30	1.14	1.84
VT	0.32	0.72	-1.36	-1.02	-2.56	1.07	-0.47

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
VA	0.35	0.46	2.99	3.33	-0.61	0.56	1.18
WA	-0.51	-1.29	-0.85	-0.56	-1.79	0.24	-0.79
WV	0.85	2.32	4.11	0.39	-0.29	1.74	1.52
WI	0.12	0.31	-2.91	-0.15	-1.64	-0.09	-0.73
WY	1.18	1.62	2.93	6.04	1.33	1.18	2.38
CENRAP	-0.02	0.39	1.11	5.46	0.24	-0.44	1.12
MANE_VU	0.34	0.37	-0.37	0.82	-1.58	0.74	0.05
MW	0.12	0.58	0.90	3.14	-0.93	1.06	0.81
VISTAS	-1.21	-0.22	4.31	9.38	1.70	-0.44	2.25

Table 5-13. Accumulated Precipitation Error (cm) by State and Time Period for the EPA 2001 Annual MM5 Simulation.

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
ALL	2.11	2.43	4.14	5.94	2.93	2.72	3.38
AL	2.93	5.71	4.25	6.24	4.02	3.51	4.44
AK	3.80	0.91	5.04	2.44	1.84	1.70	2.62
AZ	2.57	2.40	3.24	10.16	3.38	1.35	3.85
AR	4.23	2.21	5.04	9.75	3.93	3.60	4.79
CA	5.19	2.74	0.98	2.36	0.89	5.12	2.88
CO	0.90	1.68	2.96	2.80	0.88	0.76	1.66
CT	1.46	0.85	4.23	2.64	1.45	1.45	2.01
DE	1.18	1.57	3.38	7.38	0.44	2.03	2.66
FL	1.76	3.83	9.55	20.20	11.66	2.78	8.30
GA	2.07	2.80	5.19	10.62	3.76	1.50	4.32
ID	1.43	2.00	1.96	3.31	1.78	4.02	2.42
IL	1.53	1.64	3.68	6.59	3.22	2.22	3.15
IN	1.57	2.31	4.82	6.16	2.97	2.31	3.36
IA	1.12	1.85	4.18	5.80	3.15	1.13	2.87
KS	1.20	2.19	4.84	6.99	2.98	0.91	3.19
KY	1.51	2.23	8.94	14.09	3.65	3.21	5.61
LA	3.03	5.36	9.04	8.04	4.99	5.69	6.03
ME	1.29	1.76	2.45	2.79	2.59	1.15	2.00
MD	1.52	1.49	3.64	4.27	1.73	2.38	2.50
MA	0.91	2.98	3.17	2.35	1.90	0.95	2.04
MI	1.44	1.27	2.86	3.20	3.12	1.82	2.28
MN	1.22	2.58	3.50	4.08	1.95	1.08	2.40
MS	5.15	5.47	5.82	10.33	6.40	4.68	6.31
MO	2.31	2.20	4.70	7.79	3.24	1.56	3.63
MT	1.24	1.92	3.09	3.15	1.44	1.90	2.12
NE	0.95	2.06	4.93	4.45	2.24	0.82	2.57
NV	1.60	2.08	1.46	5.04	1.75	2.35	2.38
NH	2.09	2.34	2.93	2.63	3.69	1.94	2.60
NJ	1.70	2.63	2.91	3.93	1.60	1.42	2.37
NM	1.29	1.88	5.03	6.88	1.49	0.66	2.87
NY	1.37	2.02	2.74	2.89	2.55	1.56	2.19
NC	1.24	2.31	8.35	8.29	2.39	1.30	3.98
ND	0.60	0.79	2.19	2.95	1.21	0.57	1.39
OH	0.86	1.47	3.47	5.26	2.80	2.20	2.68
OK	2.53	1.52	4.96	5.83	4.07	1.69	3.43
OR	1.75	3.48	2.06	1.50	2.25	7.09	3.02
PA	0.96	2.06	2.74	3.91	2.12	1.62	2.23
RI	0.87	2.10	7.17	2.56	1.71	2.13	2.76
SC	1.67	2.89	6.74	14.40	4.62	1.20	5.25
SD	1.54	1.49	2.31	3.26	1.06	1.01	1.78
TN	3.98	1.82	5.31	13.72	3.67	4.12	5.44
TX	3.03	2.59	5.51	7.04	4.58	3.16	4.32
UT	1.68	2.01	2.81	5.20	1.82	2.29	2.63
VT	0.81	2.09	2.54	3.50	3.28	1.59	2.30

Region	Jan-Feb	Mar-Apr	May-Jun	Jul-Aug	Sep-Oct	Nov-Dec	Mean
VA	1.22	1.77	4.97	4.81	1.57	1.23	2.60
WA	2.67	3.98	2.95	2.05	3.54	7.56	3.79
WV	1.26	2.88	5.13	5.95	1.52	2.08	3.14
WI	0.76	2.04	4.41	3.44	3.04	1.12	2.47
WY	1.23	1.79	3.38	6.10	1.67	1.35	2.59
CENRAP	2.28	2.39	5.09	6.75	3.64	2.26	3.73
MANE_VU	1.25	2.08	2.86	3.30	2.42	1.55	2.24
MW	1.24	1.75	3.85	5.14	3.00	1.98	2.83
VISTAS	2.40	3.34	6.37	10.86	4.49	2.68	5.02

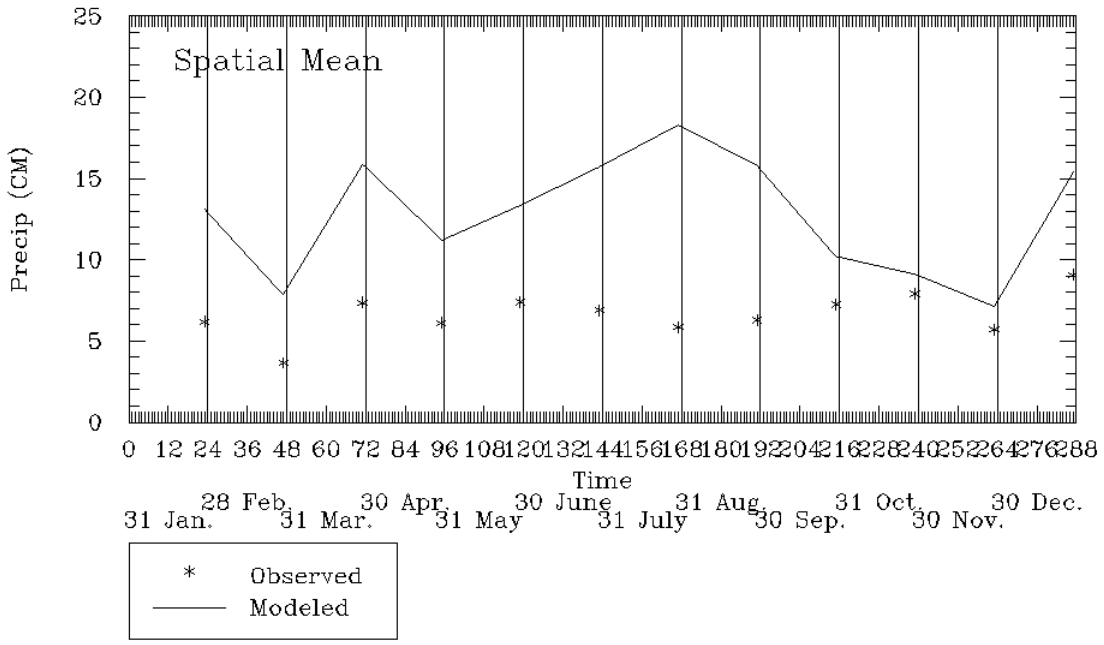


Figure 5-1: Model Estimated and Observed Mean Monthly Total Precipitation for 2001 From the WDNR Simulation Derived from CAMx Rain File.

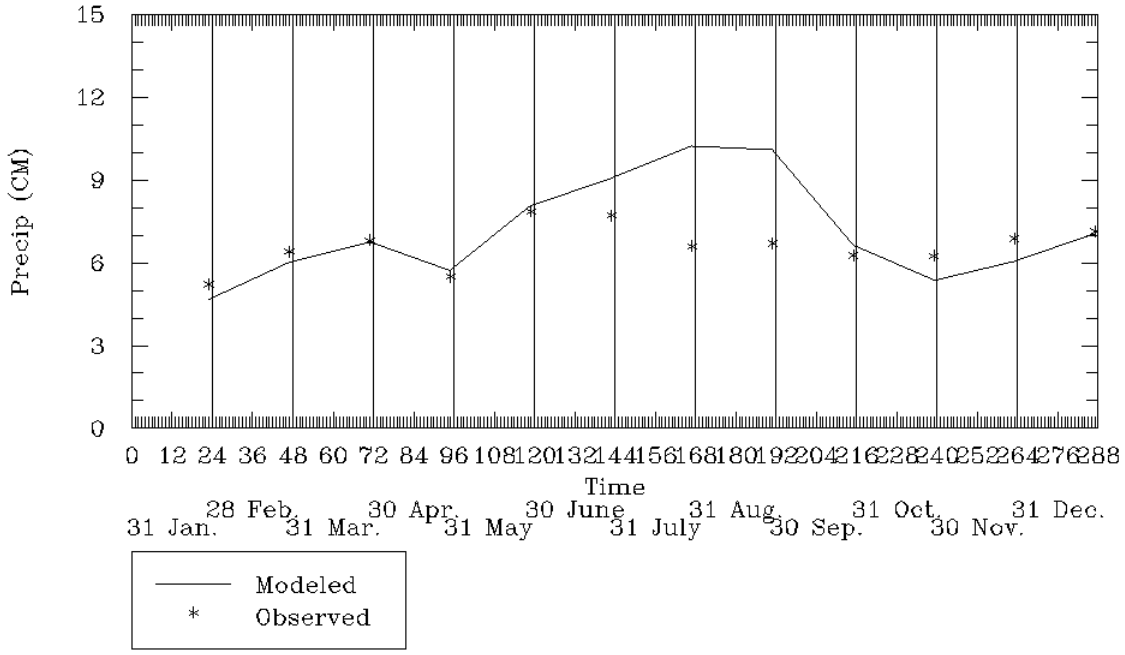


Figure 5-2: Model Estimated and Observed Mean Monthly Total Precipitation for 2001 From the EPA Simulation.

6.0 HgCAMx MODEL EVALUATION ASSESSMENT

As part of the HgCAMx model development, the ENVIRON/AER team set the modeling system up across the RPO domain and exercised it for the full 2002 annual cycle using emissions and meteorological inputs constructed by the EMS-2003 and MM5 modeling system. In this chapter, we summarize the evaluation methodology used for HgCAMx and then present our assessment of the adequacy of the evaluation. Particular emphasis is placed on the extent to which a full evaluation was performed given the extensive, and in some cases, unique data bases available for this purpose. We recognize that a number of tradeoffs and approximations were required in compiling the annual modeling data bases and in conducting the model performance evaluation. These were related to the available resources and scope of the HgCAMx model development effort and the fact that mercury measurement data bases in the study region remain relatively sparse. Thus, our comments in this chapter are aimed at assessing the completeness of the HgCAMx performance evaluations and in suggesting additional evaluative steps that might be considered by the WDNR as further experience with the HgCAMx modeling system is sought.

6.1 Components of the HgCAMx Operational Performance Evaluation

Yarwood et al., (2003a) report the evaluation of the HgCAMx model in several complimentary ways. First, the model was exercised with inert tracers to test the mass-consistency of the model and to examine the duration of the spin-up period needed in full-scale mercury simulations of the 2002 annual period. We believe that these experiments were appropriate, properly setup and exercised, and the results are consistent with our expectations and past experience with the CAMx and CMAQ models. Indeed, in inert tracer initialization experiments with the CMAQ model for the 2002 period, we found that a 2 to 3 week period was also appropriate for ensuring that the initial conditions assumed at the beginning of the simulation "washed out" of the computational domain.

6.1.1 Mercury Evaluation

The results of the annual 2002 simulation are presented in the HgCAMx final report in several qualitative and quantitative ways, specifically:

- Spatial maps of annual average Hg (II) wet and dry deposition;
- Spatial maps of annual average Hg(p) wet and dry deposition;
- Spatial difference plots for July 2002 revealing the impact of zeroing out Hg(II) formed by chemistry and deposited;
- Spatial difference plots for July 2002 revealing the impact of zeroing out Hg(II) emissions and deposition;
- Spatial difference plots for July 2002 revealing the impact of zeroing out Hg(II) lateral boundary conditions and deposition;
- Spatial difference plots for July 2002 revealing the impact of zeroing out Hg(II) top boundary conditions and deposition;
- Scatterplots of modeled vs. observed MDN annual and seasonal Hg wet deposition fluxes based on weekly average MDN measurements and model predictions;

> Annual and seasonal summaries of weekly statistics for Hg wet deposition ($\mu g/m^2$)and precipitation amounts (mm) including the average observation, average prediction, normalized bias, fractional bias, normalized error, fractional error, and r^2.

> Modeled and observed summer season total Hg wet deposition fluxes at MDN sites;

> Histograms of July 2002 total wet Hg deposition flux at two-dozen MDN sites comparing measured vs. base case and four zero-out sensitivity runs (i.e., zero chemistry, emissions, lateral BCs, and top BCs);

The spatial difference plots (Figure 4-5 in Yarwood et al.,) provide a helpful, albeit approximate, understanding of the relative contribution of emissions, chemistry, deposition, and boundary conditions (BCs) to the divalent mercury budget, at least for the month of July 2002. However, since these "zero-out" simulation results are only presented for the month of July, it is not clear to what extent the interpretations one might draw from the results hold for other months or for the annual cycle as a whole. This in not necessarily a criticism of the reporting in Yarwood et al., (2003a); indeed it is very difficult to provide for full reporting of an annual simulation of a combined MM5/HgCAMx exercise in a concise report. However, we suggest that the final report be accompanied by on or more CDs containing the full set of graphical results for all days, months, seasons and the years so that interested reviewers may peruse this 'evaluation database' to explore topics of interest.

The seasonal scatterplots of modeled versus MDN measured wet mercury deposition fluxes constitute a helpful initial screening of the model performance. These plots were developed by extracting weekly-average measurement and predictions and aggregating the results by the four seasons (summer, winter, spring, and fall). From these plots, the substantial bias in HgCAMx's overprediction of Hg wet deposition was observed. Indeed, the model overpredicted Hg wet deposition for all seasons by factors of 2 to 3 on average. The poorest performance occurred during the summertime when liquid precipitation is greatest in the study region. Subsequent sensitivity analyses confirmed persuasively that this overestimation of Hg wet deposition was largely attributable to the overestimate rainfall amounts coming from the MM5 model.

6.1.2 Ozone Evaluation

Only a very limited assessment of the HgCAMx models performance for photochemical oxidant is reported, focusing exclusively on 1-hr ozone concentrations. Indeed, the authors describe the evaluation as "…a qualitative comparison of modeled ozone levels for June 2002 to evaluate whether the modeled oxidant levels over the continental U.S. were reasonable for summer conditions". This qualitative evaluation consisted of comparing gridded maps of daily maximum 1-hr ozone on three days in June against interpolated maps of daily maximum ozone concentrations developed by EPA. The authors conclude that "Overall, the agreement for ozone was reasonable and is suitable for supporting mercury deposition modeling". No attempt is made to provide assurance that the interpolated spatial maps of 1-hr ozone are indeed suitable for comparing against the gridded model fields.

6.1.3 Fine Particulate Evaluation

The HgCAMx model's performance for fine particulate species was also examined in a qualitative fashion. Annual wet deposition maps for sulfate, nitrate and ammonium, obtained from the National Acid Deposition Program (NADP), were compared to similar maps developed from the HgCAMx output. The

observed and predicted wet depositions for sulfate, nitrate and ammonium were presented graphically for the full U.S. for spring, fall, summer and winter periods (see Yarwood et al., 2003a, Figures 4-17 through 4-28.) Using these qualitative maps (with different color and numerical scales), the authors concluded that the modeled sulfate deposition tended to be lower than observed while the modeled nitrate deposition tended to be higher than observed. Predicted ammonium deposition was much lower than observed. Overall they found that the seasonal deposition results for sulfate, nitrate and ammonium show only limited agreement with the observations for 2002. The under prediction tendencies for sulfate and ammonium are in the opposite direction to mercury suggesting sources of bias other than precipitation for sulfate and ammonium deposition. Clearly, the uncertainties in the ammonia inventory will influence ammonium deposition, but the role to which sulfate and nitrate deposition are influenced remains uncertain.

6.2 Assessment of the Present HgCAMx Evaluation

Mercury Evaluation: The HgCAMx mercury evaluation consisted of what EPA refers to as a "big picture" assessment of a regional model (EPA, 2001). Through the use of seasonal or annual averaged quantities and easy-to-interpret scatter plots, bar charts, and spatial maps (including residual plots), the broader features of the model's mercury predictions were presented. Since total wet deposition fluxes were the only mercury measurements assembled for the evaluation, these presentations naturally focused on this quantity. For the first evaluation of a new model, this cursory examination of model performance may be suitable, but it falls far short of the type of scientific evaluation needed to uncover whether the model is simulating the various chemical species correctly for the right reasons. Unlike urban ozone performance evaluations in regions such as Houston, Los Angeles or LMOS where a comparatively rich data base of precursor species measurements is available, in the Great Lakes region there are very little, if any, measurements of the three main Hg species, Hg(0), Hg(II), and Hg(p). Virtually all of the ground-level mercury data come from the annual simulated wet deposition fluxes of total mercury (Hg(0) + Hg(II) + Hg(p)) from the MDN network. While model comparisons with the MDN data sets, such as those reported by Yarwood et al., (2003a) are essential, they inherently confound the details of the atmospheric chemistry and meteorology into an aggregate quantity – wet deposition. As amply reported by the model developers, problems with the meteorological portion of the model, in this case, accurate simulation of precipitable water, confound the comparisons of modeled versus observed wet total deposition. Furthermore, lack of individual measurements of the key mercury species, (Hg(0), Hg(II), and Hg(p)), make it even more difficult to assess the adequacy of the partitioning of mercury between the oxidized and reduced forms. In sum, we believe the present Hg evaluation of the model is a useful step, but the present evaluation stops far short of testing the model across the full range of time and space scales afforded by even the limited MDN measurement network. Notwithstanding these fundamental limitations, we believe more can be done to assess the performance of the HgCAMx model. These ideas are presented in section 6.3 below.

Ozone and PM Evaluation: We find the HgCAMx evaluation for ozone and particulates to be surprisingly deficient, particularly in view of the fact that traditional statistical and graphical evaluations of the CAMx host modeling system are readily performed with available data bases (e.g., AQS, CASTnet, IMPROVE). This part of the HgCAMx evaluation almost appears as an afterthought, which is surprising since one of the key strengths of the model formulation is the direct inclusion of the VOC-NO_x-O_3 photochemical reaction pathways as well as the secondary aerosol formation mechanisms contained in CAMx4. The hourly and 24-hr gas phase and fine particulate aerosol data are readily available and could easily have been used to characterize the model's performance at least on a seasonal basis across the full annual cycle. Even though some pollutants are present in lower concentrations during winter months (e.g., ozone), it is important that the model be shown to replicate intra-annual trends. In our view, the lack of a

meaningful evaluation of the HgCAMx model's performance for ozone, NO, NO2, sulfate, nitrate, ammonium and other aerosol species constitutes an important limitation in the analysis and needs to be addressed. Demonstrating that the modeling system performs well for other precursor and product species for which substantial data – at the ground and aloft -- are available would strengthen the credibility of the new HgCAMx modeling system.

6.3 Opportunities for Extended Model Performance Evaluation

6.3.1 Ground-Level Evaluation of Photochemical and Aerosol Precursor and Product Species

There are several ways in which the current HgCAMx model evaluation can be strengthened. First, the hourly gas phase species such as NO, NO2, Ozone, and CO should be evaluated in the traditional sense (i.e., following EPA [1991] evaluation procedures) and the performance summarized on a daily, weekly, monthly and annual basis. This evaluation would cover both statistical summaries such as accuracy of peak prediction, mean normalized bias and mean normalize error (or fractional quantities) as well as graphical displays such as daily maximum tile plots, Q-Q plots, spatial mean plots, and time series at the various monitoring stations. While this represents substantial amount of plots and graphics, they need not all be presented and discussed. Simply archiving on a well organized CD would be sufficient. Similar evaluation procedures could be performed for the main aerosol species including sulfate, nitrate, ammonium, elemental carbon, organic carbon and crustal. Access to this information would allow interested analysts to explore spatial and temporal trends in the model performance that cannot be studies when the statistics are aggregated to seasonal or annual periods.

For the three mercury species, hourly, daily and seasonal tile plots, time series plots, and wet and dry deposition displays could be produced to allow analysts to study the spatial and temporal variations in the modeled response. While observational data are lacking for most of these comparisons, the presentation of the information in suitable graphical displays would allow WDNR staff to begin to acquire broader insight into the dynamic behavior of the modeling system across different time and space scales.

6.3.2 Aloft HgCAMx Performance Testing.

Evaluation of the performance of the HgCAMx model in simulating various photochemical, aerosol, and mercury species aloft is possible, largely as the result of the ongoing WDNR aircraft sampling program. While results from aircraft flights over the Great Lakes Region were not used to evaluate the model in the 2002 base case, data do exist for this purpose and the Mercury Modeling Team is encouraged to consider this extension of the HgCAMx evaluation. As part of this review, we investigated the extent to which aloft measurement data might be available to further test the HgCAMx model's predictions of precursor species, photochemical oxidants, aerosol species and mercury compounds over the Great Lake Region. A brief summary of the available data to support aloft HgCAMx evaluations are as follows.

During CY-2002, there were at least four airborne field programs underway in the eastern U.S. Two were centered over the Midwest and two mid-Atlantic coast region. The availability of these aloft data sets constitutes an additional opportunity for a scientific evaluation of the HgCAMx model evaluation using these non-routine, aloft data sets. Locally, aircraft mercury monitoring using gold trap devices (EPA Method IO05) was initiated in August 2002 and ran through the first quarter of 2003 (WDNR, 2002). Though measurements were only collected every 12th day, the data collected—largely over Lake Superior – would be helpful in corroborating the HgCAMx model estimates aloft. We were unable to

assemble additional details on the suitability of the aloft mercury data for HgCAMx performance testing within the time period of this review.

WDNR and other MPRO states have supported a comprehensive aloft sampling program for ozone and precursors over the Great Lakes Region for years. In particular, in the upper Midwest, the Lake Michigan Air Directors Consortium (LADCo) has been centrally involved in aircraft programs to support model development and applications studies for seventeen (17) years, beginning with pioneering flights in 1987. Complementing this long-term sampling performed by LADCo in the Midwest, there are other occasional intensive airborne sampling campaigns throughout the eastern U.S. (e.g., the 1999 SOS field program which provided aloft data for our evaluation of CMAQ for the July '99 episode), that have produced very useful information for air quality model performance testing.

During 2002 the Wisconsin Department of Natural Resources (WDNR) and the Midwest RPO (MRPO) collaborated on the support of airborne sampling using two aircraft that, along with ground-based measurements, provided a 3-dimensional representation of air pollution concentrations across the upper Midwest with some flight paths extending south to include the Mammoth Cave, KY and Dolly Sods, WV Class I areas. The goal of the WDNR/MROP flights was to collect aloft air quality and meteorological data to support model evaluation and data analyses. The aircraft flights were aimed at: (1) characterizing high fine particle and ozone episodes, (2) characterizing air quality over the Class I areas in the upper Midwest (Isle Royale National Park and Seney National Wildlife Refuge in northern Michigan) on both clean and hazy days, and (3) characterizing urban areas in the Midwest.

As indicated in Table 6-1, airborne sampling was performed over a broad region of the Midwest from 1 June 2002 to 22 November 2002. Lasting 3-5 hours, the WDNR and Jacko aircraft sampled a variety of aerometric parameters (depending upon the flight and aircraft) including wind speed, wind direction temperature, dew point, relative humidity, pressure, O_3, NO, NO_2, NO_x, NO_y, speciated VOCs, carbonyls, HNO_3, NH_3, Hg, SO_4, OC, EC, $PM_{2.5}$, and light scattering (Neph). Still photographs documenting visibility were also collected. Presently, the full WDNR/MRPO aircraft database, from the first flights in 1987 to the recent sampling in 2003 is being aggregated into a master data base archive.

At the University of Maryland, researchers have been using ground-based monitors, radiosondes, profilers, and instrumented aircraft to make observations each year since 1992. Parameters measured included meteorology; selected trace gases; fine particulate chemistry, microphysics and optical properties across broad regions of the middle Atlantic coast. During 2002, the University Research Foundation's Aztec-F aircraft instrument suite included O_3, NO, CO, SO_2 samplers, as well as a NO_2 closed-path tunable diode laser system, and a differential GPS-based meteorology (T, RH) and horizontal wind (*u* and *v* horizontal components) data system. Aztec-F flights were made from 23 May to 3 October, typically lasting 3 hours.

The volume of aircraft information available for HgCAMx performance testing during 2002 is quite significant and, as indicated above, some of these data include measurements of mercury species. Historically, aircraft data sets have been used only sporadically in evaluating model performance aloft. The first systematic use of such data was actually performed over the Lower Lake Michigan region in support of the Lake Michigan Ozone Study (LMOS). These aircraft data sets offer the WDNR a unique opportunity to test the new HgCAMx modeling system with an extensive aloft data base. We are unaware of any other organization active in regional or global atmospheric mercury modeling that has evaluated their models for mercury and precursor/product species using specialized aircraft data

6.4 Synthesis of HgCAMx Model Evaluation

We believe the HgCAMx model evaluation presented in Yarwood et al., (2003a) represents a useful first step in testing the overall performance of this newly developed modeling system. However, the evaluation is severely limited in scope, and beyond the useful diagnostic exercise probing the role of precipitation errors and their impact on wet Hg deposition rates, the evaluation overall lacks sufficient scope and depth to identify other potential sources of error in the various model input files and/or new model code. Strengthening the model performance testing procedures by inclusion of other gas-phase and secondary aerosol species, and possibly including performance testing aloft with data from WDNR's 2002 aircraft flights is viewed as a essential next step in building confidence that the HgCAMx tools is suitable for regulatory air quality impact analyses. The present evaluation suggests that the model is not yet suitable as a tool for supporting public decision making.

Table 6-1. Aircraft Sampling Programs Yielding Potential Data Sets for Further HgCAMx Performance Testing for CY-2002.

Aircraft Program	Meteorological Parameters & Chemical Species Measured	Sampling Program & Flight Duration	Approximate Number of Flights; Days; Aircraft
University of Maryland (UMD); Univ. Research Foundation (URF)	***Meteorology:*** WS, WD, Temp, RH, ***Air Quality***: O_3, NO, NO_2, CO, SO_2, aerosol absorption, aerosol scattering.	23 May to 3 Oct; Typically 3 hrs	54 flights, 54 days, 1 aircraft
Midwest RPO & Wisconsin DNR	***Meteorology*:** WS, WD, Temp, RH, dew point, pressure ***Air Quality***: O_3, NO, NO_2, NO_x, NO_y, speciated VOCs, carbonyls, HNO_3, NH_3, Hg, SO_4, OC, EC, $PM_{2.5}$, light scattering (Neph), visibility pictures.	1 June to 22 Nov; Typically 3-5 hrs	133 flights; 29 days; 2 aircraft (WDNR and Jacko Aircraft)

7.0 FINDINGS AND RECOMMENDATIONS

Consistent with WDNR's overall atmospheric mercury modeling system program plan, the Department commissioned an independent scientific peer-review of the HgCAMx modeling system to complement the model develop and evaluation process. Our methodology for reviewing the WDNR mercury modeling system focused on the most important chemical and physical processes. The scope of this review necessitated less on processes that are known or suspected to have little influence on model predictions. Only cursory little attention to the details of the model coding and program flow logic was possible. To optimize the resources available for this review, we built upon previous scientific peer-reviews of the various emissions, meteorological, and air quality system components of the model (e.g., Tesche et al., 1992; Kumar and Lurmann, 1997; Roth et al., 1998) and the science algorithms adopted from other models and included in the HgCAMx system (e.g., ENSR, 1993; AER, 1996; ENVIRON, 2003). Our principal findings and recommendations are presented below.

7.1 Findings

Overall, we find the HgCAMx model development and initial verification exercises carried out by ENVIRON and AER, under partial support from the WDNR, to be a credible, technically sound activity. For the specific purpose at hand – regional and local scale modeling of atmospheric mercury and its deposition products – we find the conceptual approach and technical implementation to be a logical integration of currently available state-of-science emissions, meteorological, and atmospheric modeling methods and contemporary data bases. Where we find limitations in the modeling they are, in the main, expected in a new model development program. Rarely is the first application of a complex atmospheric modeling system devoid of limitations or areas where improvements can be made. The HgCAMx modeling system, as applied to the 2002 annual data base, is no exception. Where this review has judged deficiencies or limitations to exist in the current model framework or initial applications, they derive almost exclusively from the fact that this exercise was the first formal demonstration of the new modeling system.

HgCAMx Model Formulation and Implementation

Our review has not identified any significant potential or actual weaknesses in the HgCAMx model formulation. The mercury mechanism included in the CAMx4 host model is arguably the most comprehensive chemical transformation scheme yet incorporated into a regional chemical transport model for seasonal or annual use. Particularly noteworthy is the fact that HgCAMx does not rely on estimated or interpolated fields of atmospheric oxidants (e.g., OH, O_3, H_2O_2) or particulate matter for use in the chemical mechanism but simulates these photochemical and secondary aerosol precursor and produce species from first principals. Although this adds considerable computation burden to annual simulations, and the concomitant need to compile adequate precursor inventories for ozone and PM precursor species, it yields a truly one-atmosphere modeling system that accounts for the time and space variation of the key chemical species that participate in the fate of mercury in the regional atmosphere.

Emission Inventory Development

Our review of the emission data sources used in the development of an annual mercury emissions inventory for the modeling domain of this analysis suggests that these inventories of mercury were, and still are, among the most recent and representative available given today's state-of knowledge. In our view, the 1999 emissions inventory of mercury emissions has been developed in a credible fashion and in some cases, has exceeded the quality of federally managed data. However, there are some fundamental concerns

with the model-ready emissions data sets apparently supplied to HgCAMx in this study. There is a 15% difference in the total national mercury mass between the summaries prepared by ENVIRON and WDNR. Secondly, the base emissions data pertain to 1999 while the year actually modeled was 2002. The impact of these differences, potentially enough to adversely affect the HgCAMx model verification, should be explored. Finally, recent advancements in Mexican and Canadian emission development and inventory distribution exercises have provided additional data for North American air modeling analyses. When available, these inventories and most recent U.S. data should be processed and compared to existing results.

Our review of the source categories reported in the emission inventories developed by WDNR suggest that these data are completed and comprehensive in their coverage and magnitude. We additionally recognize the extended efforts put forth by WDNR to locate, identify, and estimate alternate, unreported categories and sources of mercury emissions. We found no weaknesses with the methods and coverage of sources in the inventories prepared for modeling.

However, in reviewing the usage of the mass emissions inventory data, we believe the application of limited temporal variation to prepare CAMx input files was inappropriate for the modeling of an annual 2002 episode. In fact, the assumptions used to generate model-ready inputs for HgCAMx potentially undermine the utility of the present model performance evaluation. Contrary to the conclusion section of the ENVIRON final report, an annual emissions inventory was not used to capture the seasonal cycles in mercury deposition. In fact, only three 'representative' days from two seasons were modeled, with the remaining modeling days constructed using an unclear interpolation procedure. Thus, we believe that the actual model-ready emissions data sets input to HgCAMx may well have been inadequately representative of the collection of actual days making up the annual episode, thereby incompletely capturing the seasonal cycles of the major contributing source categories. The reliability of the HgCAMx model verification results are thus open to question.

Emissions Modeling

Overall, the emissions modeling review indicates that the mercury portion of the emissions inventory has been modeled with due care. We recommend the WDNR consider implementing two major updates to the emissions inventory modeling process in subsequent applications of HgCAMx. First, the non-mercury emissions inventory should be updated to utilize the "Base E" inventory rather than to outdated "Base D" inventory. Second, it seems appropriate to conduct a review of the default application of the speciation profiles to determine if they are appropriate. Where changes are indicated, they should be implemented before the HgCAMx modeling system is re-run with the 2002 modeling files.

Meteorological Modeling

The modeling methodology used in the HgCAMx study was compared with other recent annual modeling studies and the approach is very similar. Surface temperatures were compared with observations and while the bias and error in this application are somewhat greater than the comparison WRAP and EPA simulations, the values are close and are judged acceptable. Wind speed index of agreement scores are also somewhat poorer than the other two studies, but overall they are quite consistent with other MM5 applications for air quality studies and are deemed acceptable.

The precipitation rates used in the HgCAMx model are much higher than observations (a factor of 2-3) and the performance is markedly inferior to the MM5 performance in comparison to the EPA annual modeling for 2001. Since mercury wet deposition is proportional to rainfall rate, this large error casts

doubt on the present suitability of the HgCAMx modeling system and the 2002 episode as a credible planning tool for estimating wet mercury deposition. The model developer is well aware of this difficulty and is working to better understand the current limitations of both the MM5 meteorological model and the HgCAMx formulation. A suitable refined approach is expected. If it is critical to use the mercury wet deposition values estimated by HgCAMx, it is recommended that the rainfall rate scaled values be used. However, it is not clear whether the model's wet deposition bias is due just to precipitation problems or whether some other unknown compensating errors may be at play. According, the scaling suggestion is offered advisably and constitutes only an interim approach until more extensive model testing and evaluation is conducted.

Atmospheric Mercury Model Evaluation

The HgCAMx model evaluation presented in Yarwood et al., (2003a) represents a useful first step in testing the overall performance of this newly developed modeling system. However, the evaluation is severely limited in scope, and beyond the useful diagnostic exercise probing the role of precipitation errors and their impact on wet Hg deposition rates, the evaluation overall lacks sufficient scope and depth to identify other potential sources of error in the various model input files and/or new model code. Strengthening the model performance testing procedures by inclusion of other gas-phase and secondary aerosol species, and possibly including performance testing aloft with data from WDNR's 2002 aircraft flights is viewed as a essential next step in building confidence that the HgCAMx tools is suitable for regulatory air quality impact analyses. The present evaluation suggests that the model is not yet suitable as a tool for supporting public decision making.

7.2 Recommendations for Additional Work

Thorough peer-review of complex atmospheric modeling systems is a substantial and time consuming undertaking, one best performed by several groups working independently with a variety of modeling data bases and modeling episodes. The resources (time and budget) available to this review have limited our inspections to what we regard as the highest priority items for an initial peer review. These include an assessment of the adequacy of the model's scientific formulation, the completeness and processing of the supporting input data bases, and the quality and thoroughness of the model verification studies carried out with the full modeling system. Below, we identify additional peer-review activities that would be useful, given sufficient time and resources. Of course, some of the suggestions offered below will be implemented as a matter of course as other groups, including the WDNR Mercury Modeling Team, gain more experience and familiarity with the HgCAMx modeling system and supporting data bases.

Perform a Thorough HgCAMx Operational Performance Evaluation

The current HgCAMx model evaluation should be strengthened by conducting a thorough operational performance evaluation – at the ground and aloft – for all key gas phase and aerosol species for which observational data sets are readily available. The evaluation should consider hourly gas phase species such as NO, NO_2, O_3, and CO and the performance summarized on a daily, weekly, monthly and annual basis. For aerosol species, hourly and 24-hr gas phase and fine particulate aerosol data are readily available (e.g., AQS, IMPROVE, CASTnet) and should be employed to test model's performance on a monthly and seasonal basis across the full annual cycle. For the three mercury species, hourly, daily and seasonal tile plots, time series plots, and wet and dry deposition displays should be produced to allow analysts to study the spatial and temporal variations in the modeled response. While observational data may be lacking for some of these evaluations, the presentation of the information in suitable graphical

displays on CDs would allow WDNR staff to begin to acquire broader insight into the dynamic behavior of the modeling system across different time and space scales.

A unique opportunity is available to the WNDR with respect to aloft HgCAMx performance testing. Evaluation of the performance of the HgCAMx model in simulating various photochemical, aerosol, and mercury species aloft is possible, largely as the result of the ongoing WDNR aircraft sampling program. As noted earlier, while results from aircraft flights over the Great Lakes Region were not used to evaluate the model in the 2002 base case, data do exist for this purpose. The volume of aircraft information available for HgCAMx performance testing during 2002 is quite significant and, as indicated in Chapter 6, some of these data include measurements of mercury species. These aircraft data sets offer the WDNR a unique opportunity to test the new HgCAMx modeling system with an extensive aloft data base.

A potential hesitancy to conducting a comprehensive HgCAMx model evaluation at this time is that the emissions data base compiled to support the initial model application contained a number of simplifying assumptions that would handicap HgCAMx in performing up to its potential. However, since the WDNR's atmospheric modeling system must be viewed as a system poor HgCAMx performance for winter or spring/summer ozone and gas phase precursors for example (perhaps due to the limited representation of these periods in the emissions inventory) serves the useful purpose of identifying just how important better emissions representation is if the model is to ultimately be used as a regulatory tool for public decision making.

Strengthen the Temporal Representation of Source Emissions

For credible annual modeling of atmospheric mercury and its precursor species, we believe it is crucial to utilize a model-ready emissions inventory that possesses appropriate temporal resolution of all major sources of mercury and criteria pollutant emissions. While the current procedure of using emissions estimates from three 'representative' days from two seasons to construct a placeholder inventory for the annual cycle may be sufficient for an initial demonstration simulation of the HgCAMx model, these inputs need to be treated more realistically before the full modeling system can be used to tackle real-world regulatory or planning issues. Fortunately, established procedures are available for constructing VOC, NO_x, SO_2, CO and other pollutant inventories possessing the requisite intraannual variability and these should be used in any subsequent model applications of HgCAMx.

Additional Review of HgCAMx Model Source Code

Detailed review of a new model's source code is a costly, time consuming activity. In the case of HgCAMx, a substantial portion of the code remains the parent CAM model which has undergone detailed, albeit informal, peer-review as the result of its use by numerous public and private sector modeling groups for the past several years. Should the new mercury-specific code in CAMx4 be examined in detail, we suggest that such a code verification exercise consider the following lines of inquiry:

> Review of the mercury-specific portions of the HgCAMx computer code and model documentation to ensure that the code is consistent with the model's scientific formulation and underlying assumptions;

> Install the modeling system on additional independent computers, exercise the 2002 test case, or generate new modeling episodes to verify "transferability" of the model code and data sets;

> Continued **r**eview of the new portions of the HgCAMx source code (particularly the wet removal portion of the model) to test the structure and organization;

> Identify any platform or operating system dependent code;

> Examine the time sequencing of key operations in the chemistry and deposition routines to ensure accuracy and consistency;

> Perform 'spot checks' of new code to identify any (a) undefined or un-initialized variables, (b) inconsistent specification or dimension of arrays common to multiple subroutines, (c) inconsistencies between subroutine calls and subroutine statements, and (d) other logical errors; and

> Conduct diagnostic tests of the formulation, coding and operation of individual modules or new algorithms (e.g., daily variation in snowcover distributions);

Detailed examinations of the model source code, such as those identified above are useful and contribute to an overall strengthening of the scientific support for a new modeling system. However, we believe that given the cost of such exercises, they should be viewed as lower priority compared with another activities such as adaptation of the model to different domains and episodes (i.e., extended operational testing) to test the system under new applications.

Integration of Meteorological and Air Quality Models

The performance testing of HgCAMx against total wet deposition data from the MDN network has clearly identified the refinement of the treatment of wet removal processes as a high priority item in subsequent HgCAMx model refinement. The over-predicted surface wet deposition of total mercury has been attributed by Yarwood et al. (2003a) to several factors, the most important of which is the current approach used to derive CAMx precipitation rates from the MM5 output fields. Also, the WDNR's choice of the simple ice scheme when the MM5 model was run led to difficulties in distinguishing between liquid and frozen precipitation from MM5 when the MM5 "simple ice" scheme is used that reports only the total precipitation as liquid. An additional factor, inherent difficulties in adequately simulating the temporal and spatial patterns in observed precipitation field is, not confined to this application but has repeatedly been encountered in the application of other regional modeling systems for fine particulate and regional haze where accurate estimation of precipitation is needed (e.g., the SAMI and VISTAS regional modeling studies). It would be productive to investigate further the linkage between the nature of the output precipitation data files from MM5 (e.g., the three -dimensional instantaneous precipitation content [g/m^3] and the surface accumulated precipitation rate [mm/hr]) to assess whether some reformulation of the HgCAMx model might lead to an enhanced coupling of the two modeling systems. While the model developers have identified on option (i.e., revert to a simpler wet deposition algorithm based on the predicted surface rainfall rate or interpolated observations), we believe this approach may result in loss of physical realism in the treatment of wet deposition. An alternative is to consider exercising the prognostic meteorological model with full microphysics (notwithstanding the associated computer runtime penalties) in order to better distinguish between liquid and frozen precipitation. Still, additional investigation is needed into ways to improve consistency between the locations of clouds and rain in the meteorological simulation and the treatment in the air quality model of wet scavenging processes – at the ground and aloft. This follow-on research should be performed with one or a few well chosen wet deposition episodes rather than seasonal or annual modeling periods.

Extended Operational Testing

The HgCAMx code should be further tested focusing on subsets of the 2002 annual episode to study the models local time and space response. There would also be value in adapting the model to other episodic time periods to challenge the model with new and/or different emissions and meteorological data sets. Examining shorter duration episodes within 2002 or some other year would permit cost-effective, more highly focused diagnostic experiments to be performed. Examples of potential diagnostic examinations include: (a) sensitivity of deposition estimates to precipitation amounts and distributions, (b) sensitivity to oxidizing species and atmospheric PM, and (c) sensitivity to primary Hg emissions rates and speciation assumptions.

One of the unique and laudable features of HgCAMx is the fact that it simulates the full photochemical and secondary aerosol systems and their physicochemical interactions with elemental, divalent, and particulate mercury. Most other box and grid based atmospheric mercury models available today simply assume the ambient concentrations of SO_2, OH, H_2O_2, O_3 and other species rather than simulating them directly (CMAQ and the highly-simplified REMSAD models are exceptions). Accordingly, it would be very helpful to understand the roles that uncertainties in the assumed and/or modeled concentrations of these species have in influencing atmospheric mercury and wet and dry deposition fluxes. The HgCAMx model is a well-suited framework to address this situation in that the model can be exercised in both modes. In particular, we recommend a re-simulation of the 2002 annual period using assumed ambient concentrations of SO_2, OH, H_2O_2, O_3 and other species being used in place of the modeled values. While this may necessitate models coding changes to produce these fields rather than computing them, the expense would likely be offset by the speed with which the HgCAMx simulation would proceed. This sensitivity run would also shed light on the actual need for running the full photochemistry and secondary aerosol components of the model.

REFERENCES

AER, 1996. "Review of the UAM-AERO", prepared for the Coordinating Research Council, CRC Project No. A-19, prepared by Atmospheric and Environmental Research, Inc., San Ramon, CA.

Ariya, P.A., A. Khalizov and A. Gidas, 2002. Reactions of Gaseous Mercury with Atomic and Molecular Halogens: Kinetics, Product Studies, and Atmospheric Implications, *J. Phys. Chem.*, **106**, 7310-7320.

Bullock, O. R., and K. A. Brehme, 2002. "Atmospheric Mercury Simulation Using the CMAQ Model: Formulation Description and Analysis of Wet Deposition Results", *Atmospheric Environment*, Vol. 36, pg. 2135-2146.

Byun, D.W., and Ching, J.K.S., 1999. "Science Algorithms of the EPS Models-3 Community Multiscale Air Quality (CMAQ) Modeling System", EPA/600/R99/030. Environmental Protection Agency, Office of Research and Development, Washington, DC.

Clever, H., S.A. Johnson and E.M. Derrick, 1985. The Solubility of Mercury and Some Sparingly Soluble Mercury Salts in Water and Aqueous Solutions, *J. Phys. Chem. Ref. Data*, **14**, 631-680.

Constantinou, E. and C. Seigneur, 1993. A Mathematical Model for Multimedia Health Risk Assessment, *Environ. Software*, **8**, 231-246.

Dastoor, A. P., and Y. Larocque, 2004. "Global Circulation of Atmospheric Mercury: A Modelling Study", *Atmospheric Environment*, Vol. 38, pp. 147-161.

Doty, K., T. W. Tesche, D. E. McNally, B. Timin, and S. F. Mueller, "2002. "Meteorological Modeling for the Southern Appalachian Mountains Initiative (SAMI)", prepared for the Southern Appalachian Mountains Initiative, prepared by the University of Alabama, Huntsville, Alpine Geophysics, LLC, and Tennessee Valley Authority.

Edgerton, E.S., Hartsell, B.E.and Jansen, J.J, 2001. Atmospheric Mercury Measurements at a Rural and Urban Site Near Atlanta, GA, USA. 6th International Conference on Mercury as a Global Pollutant, 15-19 October 2001, Minamata, Japan.

ENSR, 1993. "Model Code Verification of Air Quality and Meteorological Simulation Models for the Lake Michigan Ozone Study", prepared for the Lake Michigan Air Directors Consortium, prepared by ENSR Consulting and Engineering Company, Waltham, MA.

ENVIRON, 2003. "User's Guide to the Comprehensive Air Quality Model with Extensions (CAMx)", Version 4.00. ENVIRON International Corporation, Novato, CA.

EPA, 1991. "Guidance for Regulatory Application of the Urban Airshed Model (UAM)". Office of Air Quality Planning and Standards, U.S. Environmental Protection Agency, Research Triangle Park, N.C.

EPA, 1999. "Draft Guidance on the Use of Models and Other Analyses in Attainment Demonstrations for the 8-hr Ozone NAAQS". U.S. Environmental Protection Agency, Office of Air Quality Planning and Standards, Research Triangle Park, N.C

EPA, 2001. "Guidance for Demonstrating Attainment of Air Quality Goals for $PM_{2.5}$ and Regional Haze", prepared by the EPA Office of Air Quality Planning and Standards, Research Triangle Park, NC

EPA, 2003a. "The Clear Skies Act Technical Support Package," July, 2003. http://www.epa.gov/air/clearskies/03technical_packagetofc.pdf

EPA, 2003b. "National Emission Inventory QA and Augmentation Memo", prepared by Anne Pope, U.S. EPA, Emission Factor and Inventory Group, updated June 2003.

EPRI, 2000. "Assessment of Mercury Emissions, Transport, Fate and Cycling for the Continental United States: Model Structure and Evaluation", Report No. 1000522, published by the Electric Power Research Institute, Palo Alto, CA.

Hall, B., 1995. The Gas-Phase Oxidation of Elemental Mercury by Ozone, *Water Air Soil Pollut.*, **80**, 301-315.

Hall, B. and N. Bloom, 1993. Report to EPRI, EPRI, Palo Alto, CA.

Hedgecock, I. M., and N. Pirrone, (2004). "Chasing Quicksilver: Modeling the Atmospheric Lifetime of $Hg^0_{(g)}$ in the Marine Boundary Layer at Various Latitudes", *Environmental Science and Technology*, Vol. 38, pp. 69-76.

Hetherington, 2003. "Emissions Inventory", preparation document for emissions and modeling inventories, Grant Hetherington, WDNR, November 2003.

Janssen, M. 2003a. "Use of the NEI 1999 Inventory for Regional and Continental Scale Chemical Transport Modeling of Regional Haze", Presented at 12th International Emission Inventory Conference, San Diego, CA, May 2003. http://www.epa.gov/ttn/chief/conference/ei12/modeling/janssen.pdf.

Janssen, M., 2003b. "Review of the Base E Inventory", presented at MRPO Modelers Meeting, Lake Michigan Air Director's Consortium, Des Plaines, IL, 17-18 November.

Janssen, M., 2003c. "Base E Modeling Inventory" www.ladco.org/tech/emis/BaseE/BaseE_Reports.htm.

Johnson, M. T., 2003. "Meteorological Modeling Protocol: IDNR 2002 Annual MM5 Application", Iowa Department of Natural Resources, Air Quality Bureau.

Kumar, N. and F. W. Lurmann. 1997. "Peer Review of ENVIRON's Ozone Source Apportionment Technology and the CAMx Air Quality Model", prepared for the Ohio Environmental Protection Agency, prepared by Sonoma Technology, Inc., Petaluma, CA.

LADCO. 2003. "How Was the Base D inventory Constructed." Available at the following site: http//64.27.125.175/tech/emis/BaseD/BaseD_Build.htm.

Landis, M.S. and G. Keeler, 2002. Atmospheric Mercury Deposition to Lake Michigan During the Lake Michigan Mass Balance Study, *Environ. Sci. Technol.*, **36**, 4518-4524.

Laudal, D., 2001. Final Report for JV Task 24 – Investigation of the Fate of Mercury in a Coal Combustion Plume Using a Static Plume Dilution Chamber Cooperative Agreement No. DE-FC26-98FT40321; UND Fund 4727

Lin, C.J. and S.O. Pehkonen, 1997. Aqueous-Free Radical Chemistry of Mercury in the Presence of Iron Oxides and Ambient Aerosol, *Atmospheric Environment.*, **31**, 4125-4137.

Lin, C.J. and S.O. Pehkonen, 1998. Oxidation of Elemental Mercury by Aqueous Chlorine ($HOCl/OCl^-$): Implications for Tropospheric Mercury Chemistry, *J. Geophys. Res.*, **103**, 28093-28102.

Lindqvist, O. and H. Rodhe, 1985. Atmospheric Mercury – A Review, *Tellus, Ser.B*, **37**, 136-159.

McNally, D. E., and T. W. Tesche, 2002. "Annual Meteorological Modeling Protocol (ver 1.0): Annual Application of MM5 to the Continental United States", prepared for the U. S. Environmental Protection Agency, Office of Air Quality Planning and Standards, prepared by Alpine Geophysics, LLC, Arvada, CO.

McNally, D. E., and T. W. Tesche, 2003. "Annual Application of MM5 to the Continental United States", prepared for the U. S. Environmental Protection Agency, Office of Air Quality Planning and Standards, prepared by Alpine Geophysics, LLC, Arvada, CO.

Morris, R. E., et al., 2003a, "Recent Development and Applications of the Comprehensive Air Quality Model with Extensions (CAMx)", prepared by ENVIRON International Corporations, Novato, CA.

Morris, R. E., et al., 2003b, "Recent Advances in Photochemical Air Quality Modeling Using the CAMx Model: Current Update and Ozone Modeling of Point Source Impacts", prepared by ENVIRON International Corporations, Novato, CA.

(MRPO, 2003a) Midwest Regional Planning Organization, *Base E Modeling Inventory*, September 16, 2003, http://64.27.125.175/tech/emis/BaseE/baseEreport.pdf

(MRPO, 2003b) Midwest Regional Planning Organization, *LADCO's Base E Inventory,* September 2003, http://www.ladco.org/tech/emis/BaseE/BaseE_Reports.htm

Munthe, J., 1992. The Aqueous Oxidation of Elemental Mercury by Ozone, *Atmospheric Environment, Part A*, **26**, 1461-1468.

NADP/MDN, 2003. *National Atmospheric Deposition Program (NRSP-3)/Mercury Deposition Network*, NADP Program Office, Illinois State Water Survey, 2204 Griffith Drive, Champaign, IL.

Odman, T., T. W. Tesche, and D. E. McNally, 1994. "Evaluation of CALGRID Using Two Different Ozone Episodes and Comparison to UAM Results", *Atmospheric Environment*, Vol. 28, No. 17, pp. 2823-2845.

Odman, T., J. W. Boylan, J. G. Wilkinson, A. G. Russell, 2002. "SAMI Air Quality Modeling Final Report", prepared for the Southern Appalachia Mountains Initiative (SAM), prepared by Georgia Institute of Technology, TVA, and the University of Alabama, Huntsville.

Olerud, D. 2003. "Evaluation Methodologies for Meteorological Modeling in Support of VISTAS (Visibility Improvement-State and Tribal Association), prepared for the VISTAS Technical Analysis Workgroup, NC Division of Air Quality, prepared by Baron Advanced Meteorological Systems, LLC, Research Triangle Park, NC.

Pai, P., P. Karamchandani and C. Seigneur, 1997. "Simulation of the Regional Atmospheric Transport and Fate of Mercury Using a Comprehensive Eulerian Model", *Atmospheric Environment*, **31**, 2717-2732.

Pai, P., et al., 1999. "Sensitivity of Simulated Atmospheric Mercury Concentrations and Deposition to Model Input Parameters", *Journal of Geophysical Research*, 104, D11, pp 13,855-13,868.

Pehkonen, S.O. and C.J. Lin, 1998. Aqueous Photochemistry of Divalent Mercury with Organic Acids, *J. Air Waste Manage. Assoc.*, **48**, 144-150.

Peterson, G., et al., 1998. "A Comprehensive Eulerian Modelling Framework for Airborne Mercury Species: Development and Testing of the Tropospheric Chemistry Module (TCM)", *Atmospheric Environment*, Vol. 32, pp. 829-843.

Peterson, G., et al., 2001. "A Comprehensive Eulerian Modelling Framework for Airborne Mercury Species: Model Development and Applications in Europe", *Atmospheric Environment*, Vol. 35, pp. 3063-3074.

Pirrone, N., 2001. "Mercury Research in Europe: Towards the Preparation of the EU Air Quality Directive", *Atmospheric Environment*, Vol. 35, pg. 2979-2986.

Pleijel, K., and J. Muhthe, 1995. "Modeling the Atmospheric Mercury Cycle - Chemistry in Fog Droplets", *Atmospheric Environment*, Vol 29, pp. 1441-1457.

Roth, P. M., T. W. Tesche, and S. D. Reynolds, 1998. "A Critical Review of Regulatory Air Quality Modeling for Tropospheric Ozone", prepared for the American Petroleum Institute in support of the North American Research Strategy for Tropospheric Ozone (NARSTO), Washington, D.C.

Ryaboshapko, A., et al., 2001. "Intercomparison Study of Numerical Models for Long-Range Atmospheric Transport of Mercury: Stage I. Comparison of Chemical Modules for Mercury Transformations in a Cloud/Fog Environment", Technical Report 2/2001, prepared by the Meteorological Synthesizing Center-East, Moscow, Russia.

Ryaboshapko, A., et al., 2002. "Comparison of Mercury Chemistry Models", *Atmospheric Environment*, Vol. 36, pg. 3881-3898.

Ryaboshapko, A., et al., 2003. "Intercomparison Study of Numerical Models for Long-Range Atmospheric Transport of Mercury: Stage II. Comparison of Modeling Results with Observations Obtained During Short-Term Measuring Campaigns", Technical Report 1/2003, Meteorological Synthesizing Center-East, Moscow, Russia.

Sanemasa, I., 1975. The Solubility of Elemental Mercury Vapor in Water, *Bull. Chem. Soc. Japan.*, **48**, 1795-1798.

Schroeder, W.H. and J. Munthe, 1998. Atmospheric Mercury – An Overview, *Atmospheric Environment*., **32**, 809-822.

Seigneur, C., et al., 1998. Mercury Adsorption to Elemental Carbon (Soot) Particles and Atmospheric Particulate Matter, *Atmospheric Environment*, **32**, 2649-2657.

Seigneur, C. et al., 2000. "Assessment of Mercury Emissions, Transport, Fate, and Cycling for the Continental United States: Model Structure and Evaluation", prepared for the Electric Power Research Institute, prepared by Atmospheric and Environmental Research, Inc., San Ramon, CA.

Seigneur, C. et al., 2001. "Multiscale Modeling of the Atmospheric Fate and Transport of Mercury", *Journal of Geophysical Research*, Vol. 106, No. D21, pg. 27,795-27,809.

Seigneur, C., et al., 2003a. "Scoping Study for Mercury Deposition in the Upper Midwest", AER Report CP149-03-01a, prepared for the Midwest Regional Planning Organization, Des Plaines, IL.

Seigneur, C., et al, 2003b. Modeling the Atmospheric Fate and Transport of Mercury over North America, *Fuel Processing Technol*., in press.

Sillen, G.L. and A.E. Martell, (Eds.), 1964. Stability Constants of Metal Ion Complexes, *Spec. Publ. Chem. Soc.*, **17**, 754.

Sommar, J., K. Gårdfeldt, D. Strömberg and X. Feng, 2001. A Kinetic Study of the Gas-Phase Reaction Between the Hydroxyl Radical and Atomic Mercury, *Atmospheric Environment*, **35**, 3049-3054.

Tesche, T. W., P.M. Roth, S. D. Reynolds, and F. W Lurmann, 1992. "Scientific Assessment of the Urban Airshed Model (UAM-IV)", prepared for the American Petroleum Institute, prepared by Alpine Geophysics, LLC, Crested Butte, CO.

Tesche, T. W., et al., 2001. Evaluation of CAMx and Models-3/CMAQ Over the Lower Lake Michigan Region with Inputs from the RAMS3c and MM5 Models", prepared for the Coordinating Research Council, prepared by Alpine Geophysics, LLC, and ENVIRON International Corp, Ft. Wright, KY.

Tokos, J.J.S., et al., 1998. Homogeneous Gas-Phase Reaction of Hg^0 with H_2O_2, O_3, CH_3I, and $(CH_3)_2S$: Implications for Atmospheric Hg Cycling, *Atmospheric Environment*, **32**, 823-827.

van Loon, L., E. Mader and S.L. Scott, 2000. Reduction of the Aqueous Mercuric Ion by Sulfite: UV Spectrum of $HgSO_3$ and its Intramolecular Redox Reactions, *J. Phys. Chem.*, **104**, 1621-1626.

van Loon, L.L., E.A. Mader and S.L. Scott, 2001. Sulfite Stabilization and Reduction of the Aqueous Mercuric Ion: Kinetic Determination of Sequential Formation Constants, *J. Phys. Chem.*, **105**, 3190-3195.

Vijayaraghavan, K., et al., 2002. Modeling Deposition of Atmospheric Mercury in Wisconsin, AER Report CP136-02-1, WE Energies, Milwaukee, WI.

Vijayaraghavan, K. and C. Seigneur, 2003. "Potential Impacts of the Proposed Elm Road Generating Station and of the Existing Oak Creek Power Plant on Local Mercury Deposition in Wisconsin", report

Non CP161-03-01a, prepared for WE Energies, prepared by Atmospheric and Environmental Research, Inc., San Ramon, CA.

Vijayaraghavan, K. et al., 2003. "Simulation of Mercury Deposition Over the Eastern United States with a Fine Spatial Resolution", Fourth Symposium on Air Quality: Mercury in the Atmosphere", 22-24 September, Arlington, VA.

WDNR, 2002. "Evaluation of Mercury and Nitric Acid Sampling from the Wisconsin DNR Aircraft – Fall 2002", prepared for the Aircraft Monitoring Workgroup, prepared by the Wisconsin Department of Natural Resources.

WDNR, 2003 "Hg Final Report, Emissions Inventory", provided in electronic form by Ms. Gwen Judson, WDNR.

Wesley, M. L., 1989. "Parameterization of Surface Resistances to Gaseous Dry Deposition in Regional-Scale Numerical Models", *Atmospheric Environment*, Vol. 23, jpp. 1293-1304.

Xu, X, et al., 2000a. "A Regional Scale Modeling Study of Atmospheric Transport and Transformation of Mercury. I. Model Development and Evaluation", *Atmospheric Environment*, Vol. 34, pp. 4933-4944.

Xu, X, et al., 2000b. "A Regional Scale Modeling Study of Atmospheric Transport and Transformation of Mercury. II. Simulation Results for the Northeast United States", *Atmospheric Environment*, Vol. 34, pp. 4945-4955.

Yaarwood, G. et al., 2003a. "Modeling Atmospheric Mercury Chemistry and Deposition with CAMx for a 2002 Annual Simulation", prepared for the Wisconsin Department of Natural Resources, prepared by ENVIRON International Corporation, Novato, CA., and Atmospheric and Environmental Research, Inc., San Ramon, CA.

Yarwood et al., 2003b. "Update on CAMx4 Developments", prepared by ENVIRON International Corporation, Novato, CA.